ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, U. S. ATOMIC ENERGY COMMISSION,
By ISOTOPES INFORMATION CENTER, OAK RIDGE NATIONAL LABORATORY

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Isotopes and Radiation Technology

is a Technical Progress Review prepared by the Isotopes Information Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

Insofar as feasible all references are checked by the editors, using the library facilities of Oak Ridge National Laboratory, Oak Ridge Associated Universities, and the University of Tennessee. Occasionally, however, the cited material is not available; such cases are identified by an asterisk (*).

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Heat and Power Development
Applications in Life Sciences
Safety
Isotopes Around the World
Miscellaneous

P. S. Baker, Robert H. Lafferty, Jr., Martha Gerrard, R. E. Greene, F. E. McKinney, F. J. Miller, and D. N. Hess, *Editors*
A. F. Rupp, *Adviser*

OAK RIDGE NATIONAL LABORATORY

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TECHNICAL PROGRESS REVIEWS

The United States Atomic Energy Commission publishes the Technical Progress Reviews to meet the needs of industry and government for concise summaries of current nuclear developments. Each journal digests and evaluates the latest findings in a specific area of nuclear technology and science. *Nuclear Safety* is a bimonthly journal; the other three are quarterly journals.

Isotopes and Radiation Technology
P. S. Baker, A. F. Rupp, and associates
Isotopes Information Center, Oak Ridge National Laboratory

Nuclear Safety
Wm. B. Cottrell, W. H. Jordan, J. P. Blakely, and associates
Nuclear Safety Information Center, Oak Ridge National Laboratory

Reactor Technology

Reactor Materials (Publication will be discontinued with Vol. 13, No. 3)
E. M. Simons and associates
Battelle Memorial Institute, Columbus Laboratories

All are available from the U. S. Government Printing Office. See the back cover for ordering instructions.

The views expressed in this publication do not necessarily represent those of the United States Atomic Energy Commission, its divisions or offices, or of any Commission advisory committee or contractor.

Availability of Reports Cited in This Review

United States Atomic Energy Commission (USAEC) reports are available at USAEC depository libraries and are sold by the Clearinghouse for Federal Scientific and Technical Information (CFSTI), National Bureau of Standards, U. S. Department of Commerce, Springfield, Va. 22151. All reports sold by CFSTI are $3.00 for printed copy and $0.65 for microfiche. Each separately bound part of a report is priced as a separate report. Some reports may not be available because of their preliminary nature; however, the information contained in them will generally be found in later progress or topical reports on the subject.

Other U. S. Government agency reports identified in this journal generally are available from CFSTI.

Private-organization reports should be requested from the originator.

United Kingdom Atomic Energy Authority (UKAEA) and Atomic Energy of Canada Limited (AECL) reports are available at USAEC depository libraries. UKAEA reports are sold by Her Majesty's Stationery Office, London; AECL reports are sold by the Scientific Document Distribution Office, Atomic Energy of Canada Limited, Chalk River, Ontario, Canada. UKAEA and AECL reports issued after March 1, 1967, are sold by CFSTI to purchasers in the United States and its territories.

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Section

I

Isotopes and Radiation Technology

Isotope Production and Development

Prospects for Further Considerable Extension of the Periodic Table*

By Glenn T. Seaborg

Abstract: *The possibilities of extending the periodic table to elements in the region of atomic number 114 are discussed, and the physical and chemical properties of these potential new elements, as well as of those up to number 132, are predicted. Particularly interesting is the concept of an "island of stability" surrounded by a "sea of instability," with maximum stability at approximately Z = 110, with suggested half-lives of as long as 10^8 years.*

The importance that the scientific community attaches to the formulation of the periodic classification of the chemical elements by Dmitri Mendeleev is evidenced by centennial celebrations during 1969 at places throughout the world—for example, the celebration held by the United Nations Educational, Scientific, and Cultural Organization (UNESCO) in Paris in March; a week-long symposium in Leningrad in September; a symposium in Houston, sponsored by the Welch Foundation, held in November; and a Mendeleev session at the December meeting of the American Association for the Advancement of Science, in Boston. The Houston symposium honored Mendeleev's achievement by presentation of 3 days of papers on known and predicted transuranium elements and was also the occasion for observing the 25th anniversary of the discovery of americium and curium. In all instances, scientists from countries throughout the world joined together to mark the end of one epoch of discovery and the beginning of another.

In papers previously given before the American Chemical Society and other scientific groups, I have discussed the transuranium elements, their methods of production, and their applications and have shown how the elegant simplicity of Mendeleev's periodic table led physicists and chemists to continue the search for orderliness in the chemical properties of these and other elements.[1] My own research was heavily dependent on the acceptance of periodicity among the transuranium elements. On this occasion, however, I would like to look forward—rather than backward—toward that new epoch which I believe lies ahead in extending the periodic table far beyond our myopic imagination of a few decades ago.

AN EXTENDED PERIODIC TABLE

In the modern periodic table, familiar in detail to every physical scientist and indeed in a more general sense to almost every educated person, the elements through 103 have been satisfactorily accounted for. Thus, with element 104 we enter the relatively unexplored region of the periodic system with the first member of what I have chosen to call the "transactinide" elements—that is, all elements beyond the inner transition series formed by the filling of the 5*f* electron subshell which is commonly called the "actinide series" and which includes elements 90 through 103. Once we have established the location of

*Taken from the remarks of Dr. Glenn T. Seaborg, Chairman, U. S. Atomic Energy Commission, at the American Chemical Society Mendeleev Centennial Symposium, Minneapolis, Minn., Apr. 15, 1969. A similar version has been published in *J. Chem. Educ.*, 46: 626-634 (1969). The editors wish to thank O. L. Keller, Head, Transuranium Elements Department, Chemistry Division, Oak Ridge National Laboratory, for his kind help in identifying the references cited in this article.

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the 14 elements in which the 5*f* electron shell is filled, it is possible to locate the positions of elements 104 to 121 and, in the style of Mendeleev, to predict their chemical properties with varying degrees of detail and reliability by comparing them with their homologs in the periodic table.

In Fig. 1 it can be seen that element 104 should be a homolog of hafnium, 105 a homolog of tantalum, and so forth until element 118, a noble gas homologous with radon, is reached. Using the notation of Mendeleev, I may call element 104 "eka-hafnium," element 105 "eka-tantalum," and so on. By this time your eyes will have caught my apparent brashness in extending the periodic system all the way to element 168, resulting in a periodic table of somewhat breathtaking dimensions. The most striking feature of this arrangement is the addition of another inner transition series of elements starting with about atomic number 122 and extending through atomic number 153. I call this grouping the "superactinide series," because of the rough analogy to the known actinide and lanthanide series, but hasten to point out that each element of this series does not correspond to an actinide (or lanthanide) element on a one-by-one basis. This could hardly be the case, because the superactinide series is postulated to contain 32 elements, whereas the lanthanide and actinide series contain 14 elements each. Following the superactinide transition series, elements 154 through 168 are shown as being homologous with elements 104 through 118, with element 168 again being a noble gas (or should I say a "noble liquid"?). However, it should be pointed out that calculations which are now in progress indicate a more complicated order of filling of electron shells in elements 154 through 164, which would be inconsistent with placing these elements in the periodic table in the straightforward method shown in Figs. 1 and 2.

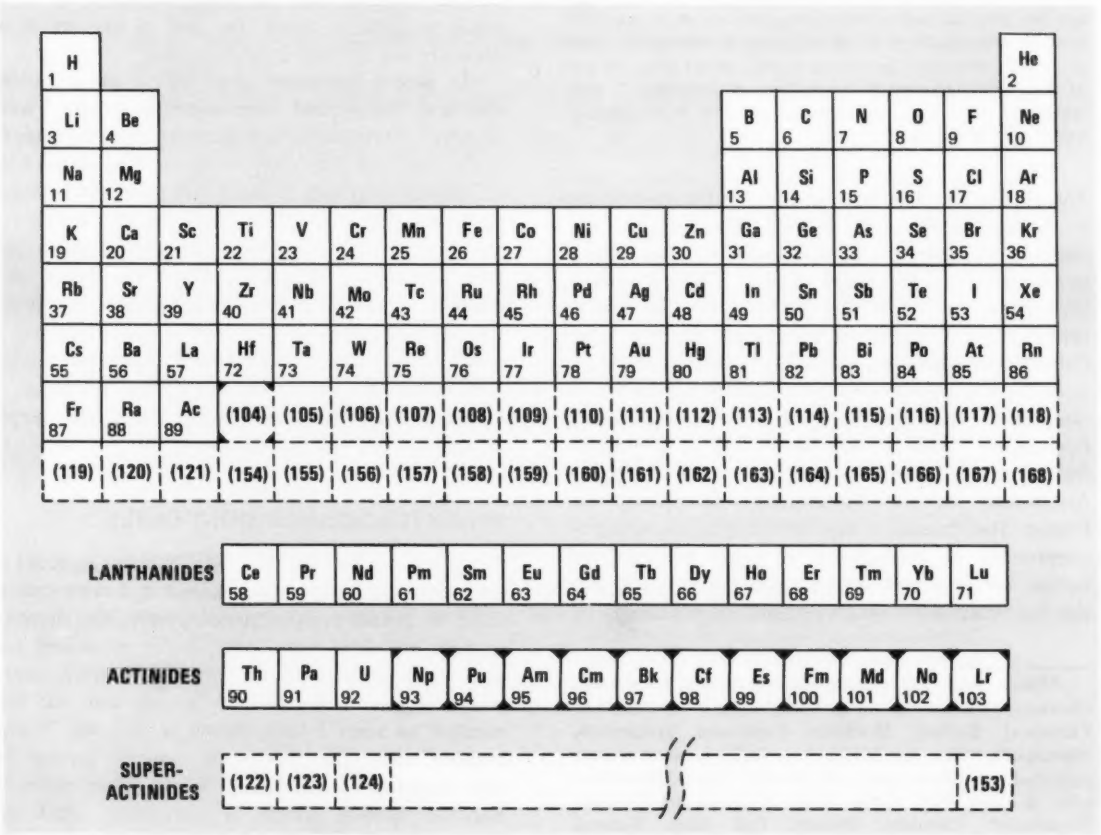

Fig. 1 Conventional form of periodic table showing predicted locations of new elements.

If this hypothetical extension of the known periodic table appears to be reckless, consider that calculations have confirmed much of it, that serious experimental efforts are being made to synthesize elements in the region around atomic number 114, and that searches for elements 110 and 114 in nature have been under way for more than a year. I will discuss these intriguing affairs in much greater detail later, but for the moment I ask acceptance for the proposition that the thought being given to what the periodic table might look like, in regions heretofore considered beyond the point of experimental verification, is much more than idle speculation.

PREDICTION OF CHEMICAL PROPERTIES OF TRANSACTINIDE ELEMENTS

Accepting the periodic classification shown in Fig. 1 as valid, I would like first to offer some predictions on the properties of the elements beyond lawrencium. Of course, these elements will first be produced only on a one-atom-at-a-time basis, and they offer scant hope for ultimate production in the macroscopic quantities that would be required to verify some of these predictions. However, many of these predicted specific macroscopic properties, as well as the more general properties predicted for the other elements, will be useful in designing tracer experiments for the chemical identification of any of these elements which might be synthesized according to the considerations to be described a little later.

Element 104 (eka-hafnium) is predicted to resemble its homolog, hafnium (element 72), in its chemical properties; thus it should also resemble thorium, but somewhat less. It should be predominantly tetrapositive both in aqueous solution and in its solid compounds, although it should form solid halides of the (II) and (III) oxidation states as well.

One should be able to predict some of the crystallographic properties of tetrapositive element 104 by extrapolation from those of its homologs, zirconium and hafnium. The ionic radii of tetrapositive zirconium (0.74 A) and hafnium (0.75 A) suggest an ionic radius of about 0.78 A for tetrapositive element 104, allowing for the smaller actinide than lanthanide contraction. Since the ionic radius of tetrapositive thorium is 0.98 A, one would expect element 104 to have a crystal chemistry like that of hafnium and quite different from that of thorium. Further, one would expect the hydrolytic properties of element 104 to be similar to those of hafnium. The sum of the ionization potentials for the first four electrons should be less than that for hafnium, which suggests that it should be easier to oxidize element 104 to the (IV) state. Some of these and other properties of element 104 are tabulated in Table 1, which has been prepared with the assistance of O. L. Keller, Jr., and J. L. Burnett of Oak Ridge National Laboratory (ORNL).

Table 1 Some Predicted Properties of Element 104

	Element 104 (eka-hafnium)
Atomic weight	272
Atomic volume, cm^3/mole	14
Density, g/cm^3	18
Most stable oxidation state	+4
Oxidation potential, volts	$M \rightarrow M^{4+} + 4e^-$ > + 1.7
Ionic radius, A	0.78
Crystal structure	hcp
Metallic radius, A	1.6
Melting point, °C	2100
Boiling point, °C	5500

Element 105 (eka-tantalum) should resemble tantalum and niobium, and to a lesser extent protactinium, with the pentavalent state being perhaps the most important. The chemical properties of element 106 (eka-tungsten) should be similar to those of tungsten, molybdenum, and to some extent chromium. Element 107 should be an eka-rhenium and element 108 an eka-osmium, which suggests that the latter should have a volatile tetroxide that should be useful in designing experiments for its chemical identification. Elements 109, 110, and 111 (eka-iridium, eka-platinum, and eka-gold) should be noble metals. If the upper oxidation states are stable, volatile hexa- or octafluorides might be useful for chemical separation purposes. As I shall describe later, there is a special interest in element 110 and its expected eka-platinum character. Element 112 (eka-mercury), because of the distinctive chemical properties of its homolog, mercury, should have properties that can be exploited in its chemical identification.

There is special interest in the properties of elements in the vicinity of atomic number 114. With the assistance of O. L. Keller, Jr., J. L. Burnett, T. A. Carlson, and C. W. Nestor, Jr., of ORNL, I have tabulated some predicted properties of elements 113 and 114, eka-thallium and eka-lead, respectively

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(Table 2). Details of the methods of extrapolation employed to arrive at the values shown will appear in the literature[2] but are too exhaustive to describe at this time. However, I would like to dwell for a moment on the oxidation states and oxidation potentials of these two hypothetical elements because they are of particular interest to me.

Table 2 Some Predicted Properties of Elements 113 and 114

	Element 113 (eka-thallium)	Element 114 (eka-lead)
Chemical group	III	IV
Atomic weight	297	298
Atomic volume, cm^3/mole	18	21
Density, g/cm^3	16	14
Most stable oxidation state	+1	+2
Oxidation potential, volts	$M \rightarrow M^+ + e^-$ -0.6	$M \rightarrow M^{2+} + 2e^-$ -0.8
First ionization potential, ev	7.4	8.5
Second ionization potential, ev		16.8
Ionic radius, A	1.48	1.31
Metallic radius, A	1.75	1.85
Melting point, °C	430	70
Boiling point, °C	1100	150
Heat of vaporization, kcal/mole	31	9
Heat of sublimation, kcal/mole	34	10
Debye temperature, °K	70	46
Entropy, eu/mole (25°C)	17	20

The Group IV elements show increasing stability in the (II) oxidation state relative to the (IV) state as one goes to higher atomic numbers. Carbon and silicon have very stable tetrapositive oxidation states, and germanium shows a very unstable dipositive oxidation state, in addition to a stable tetrapositive state. In tin, both the dipositive and tetrapositive oxidation states are important, and lead is most stable in the (II) oxidation state. In a publication[3] in 1958, R. S. Drago showed from thermodynamic considerations that this trend arises from a decrease in the strength of the covalent bonds formed by the metal atom as the atomic number increases in Group IV. From the point of view of valence-bond theory, the tetrapositive state is made possible by sp^3 hybridization, which involves all four outer electrons in bonding. Thus the tetrapositive state requires strong enough covalent bonds to supply both the promotion energy for one *s* electron and the Gibbs free energy required for compound stability. In lead, weak covalent bonds, involving only the *p* electrons, tend to be formed.

Drago explained this tendency toward involvement of only *p* electrons with increasing *Z* by noting that (1) the heavier elements have more inner electrons to repel the inner electrons of the bonded anion and (2) the valence electrons in the higher *Z* elements are spread over a larger volume so that there is less overlap with the orbitals of the anion.

Since element 114 would have the largest atomic volume and the most inner electrons of the Group IV elements, sp^3 hybridization would probably be very unimportant, and thus this element would be weakly, if at all, tetrapositive; hence the most stable oxidation state is expected to be the (II) state. The oxidation potential for the reaction

$$114 \rightarrow 114^{2+} + 2e^-$$

is calculated by Keller et al.[2] to be −0.8 volt, on the scale where the reaction

$$\tfrac{1}{2}H_2 \rightarrow H^+ + e^-$$

is assigned the value 0.0 volt.

For reasons similar to those which suggest a stable (II) oxidation state in element 114, element 113 (a member of Group III) is expected to have a preferred oxidation state of (I); an oxidation potential of −0.6 volt is predicted.

Certain predicted volatility characteristics of elements 115, 116, 117, and 118 (eka-bismuth, eka-polonium, eka-astatine, and eka-radon) or their compounds may offer advantages for chemical identification; this, of course, is especially true for element 118.

Elements 117 to 120 lend themselves to fairly detailed predictions of their macroscopic properties. B. B. Cunningham of the Lawrence Radiation Laboratory, University of California, Berkeley, and O. L. Keller, Jr., and J. L. Burnett of ORNL have made estimates of the properties of elements 117, 119, and 120 (Table 3); the properties of element 118 are those predicted by A. V. Grosse[4] in 1965. These properties are obtained by extrapolation of known properties of lower homologs in the periodic table.

The atomic weights given in Tables 1, 2, and 3 correspond to the mass numbers of the isotopes predicted to have the longest overall half-lives, taking into account the possibilities for decay by spontaneous

Table 3 Some Predicted Properties of the Heavier Transactinides

	Element 117 (eka-astatine)	Element 118 (eka-radon)	Element 119 (eka-francium)	Element 120 (eka-radium)
Chemical group	Halogen	Noble gas	Alkali metal	Alkaline earth
Atomic weight	311	314	315	316
Atomic volume, cm^3/mole	45	50	80–90	45
Density, g/cm^3			3	7
Oxidation potential, volts	$2M^- \rightarrow M_2 + 2e^-$ +0.25–0.5		$M \rightarrow M^+ + e^-$ +2.9–3.0	$M \rightarrow M^{2+} + 2e^-$ +2.9
First ionization potential, ev	9.3	9.8	3.4–3.8	5.4
Second ionization potential, ev	16	15	23	10
Electron affinity, ev	2.4–2.6			
Ionic radius, A	2.3		1.8–1.9	1.5–1.7
Crystal structure			bcc	bcc
Metallic radius, A			2.9	2.3
Covalent radius, A	1.8	2.3		
Melting point, °C	350–550	−15	0–30	680
Boiling point, °C	610	−10	630	1700

fission and by alpha-particle and beta-particle emission. These atomic-weight estimates do not affect the prediction of chemical properties and are given in order to meet the formal requirement for assignment of an atomic weight. The basis for these estimates will be described later; in some cases these are of a very preliminary nature and are surely subject to change.

ELECTRONIC STRUCTURE OF TRANSACTINIDE ELEMENTS

The electronic configurations of the elements, of course, determine their positions in the periodic table. In Fig. 2, which is a form of the periodic table that shows the filling of electron shells, it can be seen that the lanthanide series of elements, cerium through lutetium, is a transition series formed by the sequential filling of a 14-member inner electron ($4f$) subshell. It is for this reason that the chemical properties of the lanthanides are similar—but not identical. In analogous fashion, the actinide series is formed by the filling of the inner $5f$ electron subshell, with each element being somewhat similar in properties to its lower lanthanide homolog. The 14-member actinide series is completed with element 103, as was noted earlier. Substantiation of the actinide concept has come from determination of the chemical properties of the actinide elements and from various kinds of spectroscopic measurements of their electronic energy levels.

Although exceedingly complex and voluminous calculations are involved, it is possible, in principle, to calculate the electronic structures of the actinide elements—and of the undiscovered transactinide elements—well into the region of the superheavy elements.[5] Modern high-speed computers now make such calculations feasible, and they have been carried out by J. T. Waber, D. T. Cromer, D. Liberman, A. C. Larson, J. B. Mann, R. D. Cowan, and D. C. Griffin at the Los Alamos Scientific Laboratory for elements as high as atomic number 132. Their ground-state electronic configurations for elements 104 to 121 are summarized in Table 4. These electronic structures are consistent with the arrangement of transactinide elements shown in Figs. 1 and 2. Elements 104 to 112 are formed by the filling of the $6d$ subshell, making them homologous in chemical properties with the elements

Table 4 Calculated Electronic Ground States for Gaseous Transactinide Elements 104–121

Atomic No.	Electronic structure*	Atomic No.	Electronic structure†
104	$6d^2 7s^2$	113	$7s^2 7p^1$
105	$6d^3 7s^2$	114	$7s^2 7p^2$
106	$6d^4 7s^2$	115	$7s^2 7p^3$
107	$6d^5 7s^2$	116	$7s^2 7p^4$
108	$6d^6 7s^2$	117	$7s^2 7p^5$
109	$6d^7 7s^2$	118	$7s^2 7p^6$
110	$6d^8 7s^2$	119	$7s^2 7p^6 8s^1$
111	$6d^9 7s^2$	120	$7s^2 7p^6 8s^2$
112	$6d^{10} 7s^2$	121	$7s^2 7p^6 7d^1 8s^2$

*In addition to a "core" of radon, element 86, plus $5f^{14}$

†In addition to a "core" of radon, element 86, plus $5f^{14} 6d^{10}$.

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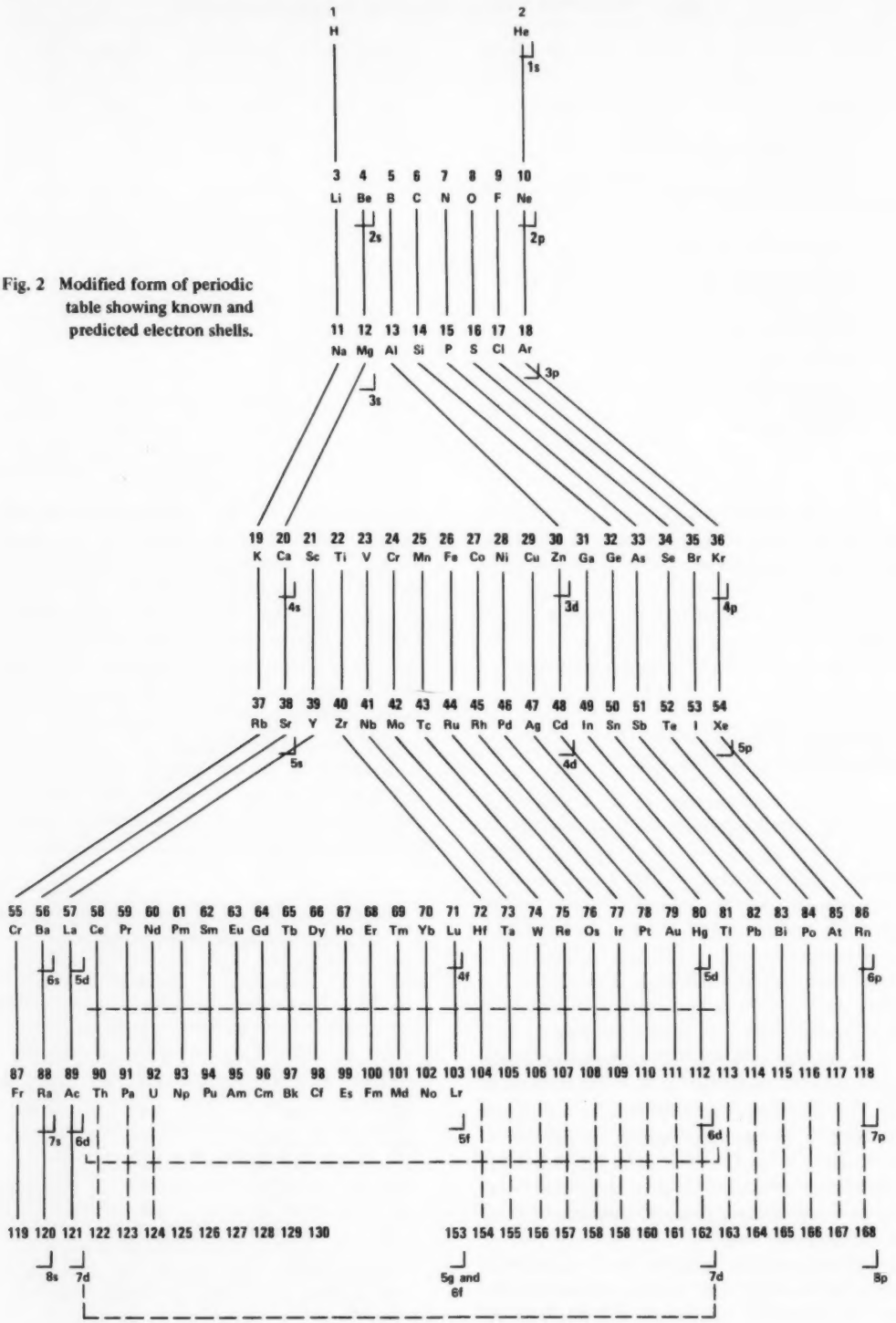

Fig. 2 Modified form of periodic table showing known and predicted electron shells.

hafnium through mercury. Elements 113 through 118 result from the filling of the 7*p* subshell and are thus similar to the elements thallium through radon. Element 119 should be an alkali metal, and element 120, an alkaline earth, formed by addition of electrons to a new 8*s* subshell. Element 121 should be similar in properties to actinium and lanthanum and results from the addition of the first electron to a new 7*d* subshell.

Several of us,[5] including the Soviet chemist V. I. Goldanskii, have speculated for some years that another inner-transition series of elements, somewhat like the lanthanide and actinide series, should begin somewhere around element 120. Quantum theory indicated that this series would be formed by the addition of 18 electrons to an inner "*g*" subshell (the "*g*" notation appearing here for the first time in the periodic system) or of 14 electrons to an inner 6*f* subshell, but the order of filling these shells could not be predicted until the Los Alamos computer techniques became available. The calculations suggest (Table 5) that, after the addition of some electrons to the 7*d* and 6*f* subshells, the filling of the 5*g* subshell takes place in an orderly manner. More recent calculations by Mann at Los Alamos now indicate the involvement of a new 8*p* subshell as well as of the 7*d* and 6*f* subshells in the early part of this transition series (Table 6). It is reasonable to expect that the filling of the inner 5*g* subshell would be followed by the filling of the inner 6*f* subshell, leading to an inner-transition series of 32 elements ending with element 153. The lower members of the series might be generally homologous with the lower members of the actinide series. Throughout the series, however, the correlation would be indistinct, although the tripositive oxidation state might be the distinctive one. Since the difference in energy levels of successive electrons is very small, this superactinide series of elements will exhibit multiple, barely distinguishable oxidation states, leading to very complicated chemistry. (More strictly speaking, apparently the elements 140 through 153 could be called the "eka-actinide" elements.)

Table 5 Calculated Electronic Ground States for Gaseous Superactinide Elements 121–132

Atomic No.	Electronic structure*	Atomic No.	Electronic structure*
121	$7d^1\,8s^2$	127	$5g^3\,6f^3\,7d^1\,8s^2$
122	$5g^0\,6f^0\,7d^2\,8s^2$	128	$5g^4\,6f^3\,7d^1\,8s^2$
123	$5g^0\,6f^1\,7d^2\,8s^2$	129	$5g^5\,6f^2\,7d^2\,8s^2$
124	$5g^0\,6f^3\,7d^1\,8s^2$	130	$5g^6\,6f^2\,7d^2\,8s^2$
125	$5g^1\,6f^3\,7d^1\,8s^2$	131	$5g^7\,6f^1\,7d^3\,8s^2$
126	$5g^2\,6f^3\,7d^1\,8s^2$	132	$5g^8\,6f^1\,7d^3\,8s^2$

*In addition to a "core" of element 118.

Table 6 More Recent Electronic Ground States for Gaseous Superactinide Elements 121–126

Atomic No.	Electronic structure*
121	$8p^1\,8s^2$
122	$5g^0\,6f^0\,7d^1\,8p^1\,8s^2$
123	$5g^0\,6f^1\,7d^1\,8p^1\,8s^2$
124	$5g^0\,6f^3\,7d^0\,8p^1\,8s^2$
125	$5g^1\,6f^3\,7d^0\,8p^1\,8s^2$
126	$5g^2\,6f^2\,7d^1\,8p^1\,8s^2$ or $5g^2\,6f^3\,8p^1\,8s^2$ (tentative)

*In addition to a "core" of element 118.

After the completion of the superactinide series, the addition of electrons to the remaining positions in the 7*d* subshell would form elements 154 through 162—i.e., eka-104 through eka-112. Filling the 8*p* subshell of six electrons would result in elements 163 through 168.

ELEMENTS 104 AND 105

Let us now examine what has been done experimentally to extend the periodic system to the region immediately beyond the previously accepted upper boundary of known elements at element 103 and to outline the prognosis for making a major advance in discovering much heavier (superheavy) elements.

Last spring, Albert Ghiorso and colleagues[6] announced that conclusive proof of the discovery of two isotopes of element 104 has been obtained. Soviet workers have discussed previously their less conclusive evidence for the discovery of an isotope of element 104 and two isotopes of element 105.

The significance of the work on elements 104 and 105 requires a brief review of the nature of the Soviet and American research and the results that have been obtained. In 1964, Flerov and his coworkers[7] published results obtained by use of the 310-cm cyclotron at the Joint Institute for Nuclear Research, Dubna, USSR, with the postulated nuclear reaction:

$$^{242}_{94}\mathrm{Pu} + {}^{22}_{10}\mathrm{Ne} \rightarrow {}^{260}104 + 4\,{}_0n^1$$

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The measured half-life of the product isotope was only about 0.3 sec, with decay occurring solely by spontaneous fission, a nonspecific decay mechanism that, contrary to alpha-particle decay, does not characterize an isotope with an identifying, precisely measurable energy. Zvara and colleagues[8] in the Soviet group have reported experiments aimed at separating this product from the other materials present by a technique based on the premise that element 104 would be the first transactinide element and hence would have chemical properties similar to those of hafnium, below which it would fall in the periodic table (see Figs. 1 and 2). The technique consisted in attempting to convert any product atoms to chlorides and making use of the fact that the tetrapositive elements of Group IV (hafnium and zirconium) have very volatile chlorides when compared with the relatively nonvolatile chlorides of the actinide elements in the (III) oxidation state. They concluded that element 104 indeed showed the properties of a Group IV element; that is, they believed that they had observed the 0.3-sec spontaneous fission activity in the volatile tetrachloride fraction. The Soviet scientists have been using the name "kurchatovium" (symbol Ku) for the new element they believe that they have observed.

However, the difficulty of performing accurate chemical experiments when the half-life is so short, and the existence in the actinide region of numerous short-lived isomers that decay by spontaneous fission, leads to the possibility of confusion and incorrect assignment of atomic number and emphasizes the need for independent confirmatory experiments. In fact, attempts beginning in 1967 by Ghiorso and associates at Berkeley to produce an isotope of element 104 with the same properties reported by the Soviet workers—decay by spontaneous fission with a half-life of 0.3 sec—have been unsuccessful so far. On the other hand, the Berkeley group[6] (A. Ghiorso, M. J. Nurmia, J. A. Harris, and P. L. and K. A. Y. Eskola), in experiments performed in 1968 and 1969, has bombarded the californium isotope ^{249}Cf with ^{12}C and ^{13}C ions in the Heavy Ion Linear Accelerator (HILAC), expecting that the following nuclear reactions might occur:

$$^{249}\text{Cf} + {}^{12}\text{C} \rightarrow {}^{257}104 + 4n$$

$$^{249}\text{Cf} + {}^{12}\text{C} \rightarrow {}^{258}104 + 3n$$

$$^{249}\text{Cf} + {}^{13}\text{C} \rightarrow {}^{259}104 + 3n$$

The first and third reactions result in isotopes that decay by alpha-particle emission with half-lives of about 4 sec and alpha energies in the range 8.7 to 9.0 Mev, and the second reaction results in an isotope that decays by spontaneous fission with a half-life of about 0.01 sec. As confirmatory evidence for $^{257}104$ and $^{259}104$, they observed as their decay products the previously known isotopes $^{253}102$ and $^{255}102$; this constitutes definite proof for the identification of the two alpha-particle-emitting isotopes $^{257}104$ and $^{259}104$. More recently Ghiorso and his group have bombarded ^{248}Cm with ^{18}O and produced $^{261}104$, which decays by the emission of alpha particles with a half-life of about 1 min; this was proved by observing as its decay product the previously known $^{257}102$. Use of the isotope $^{261}104$ should make it possible to study the chemistry of element 104.

There is some very preliminary information on isotopes which could conceivably be assigned to the element with atomic number 105. Flerov and his coworkers have described the observations they made in 1967 as the result of the bombardment of ^{243}Am with ^{22}Ne ions in the 310-cm cyclotron at Dubna. They observed alpha-particle emitters with energies of 9.7 ± 0.1 and 9.4 ± 0.1 Mev and half-lives greater than 0.01 sec (and between 0.1 and 3 sec) which they attribute to $^{260}105$ and $^{261}105$. The observed yield is 0.05 atom/hr. The identification depends on the correlation of the alpha-particle decay with daughter alpha-particle emitters of energy 8.6 Mev, which they postulate as $^{256}103$ and $^{257}103$. Again the efforts of Ghiorso and coworkers to confirm these experiments have been unsuccessful so far.

PREDICTED NUCLEAR STABILITY OF SUPERHEAVY ELEMENTS

With regard to the development of theories of nuclear stability of the undiscovered, very heavy (superheavy) elements, we must go back 20 years, when Maria Goeppert Mayer[9a] and O. P. L. Haxel, J. H. D. Jensen, and H. E. Suess[9b] began to develop a theoretical "shell" model of the nucleus which consisted of a collection of particles moving in a nuclear force field. The collection of particles—neutrons and protons—was shown to be especially stable when the nucleus contained a "magic" number of neutrons or protons, or, more simply, nuclear shells similar to the extranuclear electron shells of the Bohr atom. Magic numbers of neutrons (N) or protons (Z) are generally recognized as being 2, 8, 20, 28, 50, and 82 in the elements below uranium in the periodic table. The magic number $N = 126$ is significant in this region also, as can be seen from the special stability of ^{208}Pb

($Z = 82$, $N = 126$), a doubly magic, spherically symmetrical nucleus. This shell theory has progressed through many improvements to the point where the potentials of single nucleons in a deformed or non-spherical nuclear field can be calculated, using the now widely accepted method of the Swedish physicist Sven Gösta Nilsson[10]—the so-called "Nilsson orbitals."

The well-known charged liquid-drop model of the nucleus, which was so effective in developing the theory of neutron-induced nuclear fission, also has been developed further to help in predicting stabilities of heavy nuclei toward decay by spontaneous fission and other modes. In the extension of the liquid-drop model formulated by W. D. Myers and W. J. Swiatecki,[11] the potential energy of the nucleus is a function of neutron number, atomic number, and nuclear shape, and a shell correction is applied when the shape approaches sphericity. The semiempirical mathematical expressions which describe or are related to this model can be used to calculate ground-state masses and equilibrium deformations of hypothetical nuclei as functions of N and Z, as well as the barrier energies and related half-lives for spontaneous fission. Most isotopes, including those of the lanthanide and actinide elements, have more or less deformed (such as ellipsoidal) shapes, but those in the immediate vicinity of closed nucleon shells have spherical shapes. In the absence of closed shells, all superheavy elements would decay by spontaneous fission with half-lives so short as to preclude their production and observation.

Recently, exciting theoretical calculations, made by scientists from many countries, including Nilsson, V. M. Strutinskii, C. Gustafson, S. A. E. Johansson, H. Meldner, Swiatecki, F. A. Gareev, and P. A. Seeger, have used the liquid-drop model with shell corrections taken from Nilsson orbitals—using extrapolations of known parameters from the lanthanide and actinide regions—to establish the possible existence of closed nucleon shells in the region of superheavy elements. The survival of such nuclei from decay by spontaneous fission means that their rate of decay by alpha- and beta-particle emission becomes important; and therefore estimates of stability toward these modes of decay have also been made by Nilsson and his coworker, Chin-Fu Tsang, at Berkeley.[12a]

Such theoretical considerations suggest the existence of closed nucleon shells at $Z = 114$ and $N = 184$, which exhibit great stability against decay by spontaneous fission. Fortunately the nucleus with both $Z = 114$ and $N = 184$ is situated near the bottom of the potential energy valley of stability; that is, it is stable against decay by beta-particle emission and is doubly magic just like beta-stable ^{208}Pb. The ^{298}114 nucleus is, of course, unstable toward alpha-particle decay. A less stable closed shell has been computed in the past to exist at $Z = 126$ (or $Z = 124$), but this is no longer considered likely. The calculations indicate the existence of a closed shell at $N = 196$ and another closed nucleon shell may occur at $Z = 164$. These calculations are based on the Nilsson model and must still be confirmed by calculations with other models such as the Woods-Saxon potential.

As was indicated earlier, element 114 might be termed "eka-lead" by use of Mendeleev's nomenclature because it is a homolog of lead in the periodic table; likewise element 164 would be "eka-114" or "eka-eka-lead." It is especially interesting to note that the known and predicted centers of nuclear stability—atomic numbers 50, 82, 114, and 164—fall in the same column (Group IV) of the periodic table. This is curious because the principles that determine nuclear stability are apparently not related to those which determine electronic structure (which determines an element's position in the periodic table).

Enhancing the prospects for the actual synthesis and identification of superheavy nuclei is the fact that the doubly magic nucleus ^{298}114 is actually the center of a rather large region of stability, with maximum stability against overall decay by all processes (spontaneous fission and alpha- and beta-particle emission) occurring at $Z = 110$, $N = 184$ (that is, ^{294}110) and with decreasing stability being found as the edges of the region are reached. This is represented in an allegorical fashion in Fig. 3, which I have prepared with the assistance of W. J. Swiatecki. (For the purposes of dramatization, this allegory reverses the usual method of display of centers of stability which are conventionally depicted as valleys in the potential energy surface.) The long mainland peninsula corresponds to the whole region of stable nuclei and the "island of stability" corresponds to the region of predicted relative stability among the superheavy elements. Surrounding these is a "sea of instability." The grid lines (at sea level) represent magic numbers of protons (Z) or neutrons (N). The doubly magic region on the mainland peninsula at $Z = 82$, $N = 126$ is represented by a mountain. Other areas of enhanced stability on the peninsula due to single magic numbers are shown as ridges. The region between $Z = 83$ and $Z = 90$ is shown as being at a somewhat lower elevation (but above sea level) than the other parts of the peninsula of stability because of the appearance of alpha-particle emission as a predominant mode of decay. Submerged ridges indicate that the isotopes are unstable but are relatively

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more stable than the neighboring ones because of the increased stability afforded by closed nucleon shells. The island of stability has an irregular shape reflecting the doubly magic region (centered at $Z = 114$, $N = 184$) and the closed shell at $N = 196$, and the rate of decay is determined by competition between spontaneous fission and alpha- and beta-particle emission. This island also reflects the maximum in overall half-life predicted for the vicinity of $Z = 110$.

stability in the region of interest are shown in Fig. 4. Quantitatively the numbers may be in error by orders of magnitude, but, even if the actual values are lower than indicated by many factors of 10, any produced nuclei that have the appropriate atomic number and mass number should still be observable. The doubly magic nucleus $Z = 114$, $N = 184$, for example, is predicted to have a spontaneous-fission half-life of 10^{16} years and an alpha-decay half-life of greater than

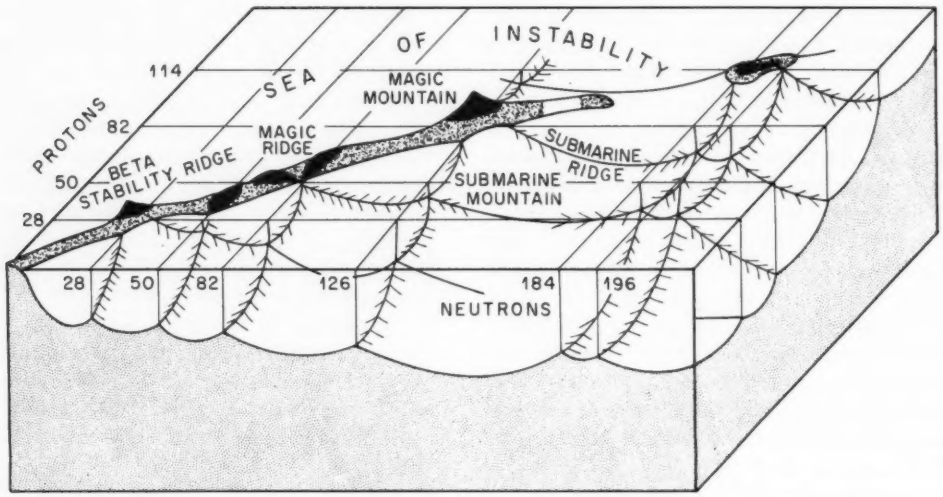

Fig. 3 Known and predicted regions of nuclear stability, surrounded by a sea of instability.

Now that I have demonstrated where the region of enhanced nuclear stability lies by use of allegorical geography, I would like to present the results of the scientific computations which define the region in parameters of half-life values, atomic number, and neutron number.

If the nuclei in this island of stability are to be observed, they must have favorably slow rates of decay by alpha- and beta-particle emission, which are independent of and determined by different considerations than those which determine decay by spontaneous fission. Some nuclei are predicted to be stable against beta decay, some might have half-lives as long as 10^{13} years for decay by alpha-particle emission and some perhaps 10^{16} years for decay by spontaneous fission, although no specific nucleus would exhibit all of these favorable properties simultaneously! The actual trends predicted by Tsang[12] for spontaneous-fission and alpha and beta half-life values and beta

l year. My own estimate is that the alpha-decay half-life may approach 10^3 years. This nucleus should be stable against beta-particle decay. Half-lives of the order of seconds, even down to nanoseconds, are readily discernible with modern instrumental methods used in detection of new elements, so there is quite a margin of allowable error in the use of the predictions to select a region for experimental study. Also, the spread of predicted half-lives means that, if the island of stability can be entered experimentally, the investigator should be rewarded by the production of an isotope (or isotopes) with half-life suitable for detection.

Since the data that have been accumulated by Tsang and presented in Fig. 4 are somewhat difficult to assess in a concise manner, I have chosen to narrow the discussion of the region of stability by taking two cross sections of the region, one for $Z = 114$ constant (Fig. 5) and the other for $N = 184$ constant (Fig. 6),

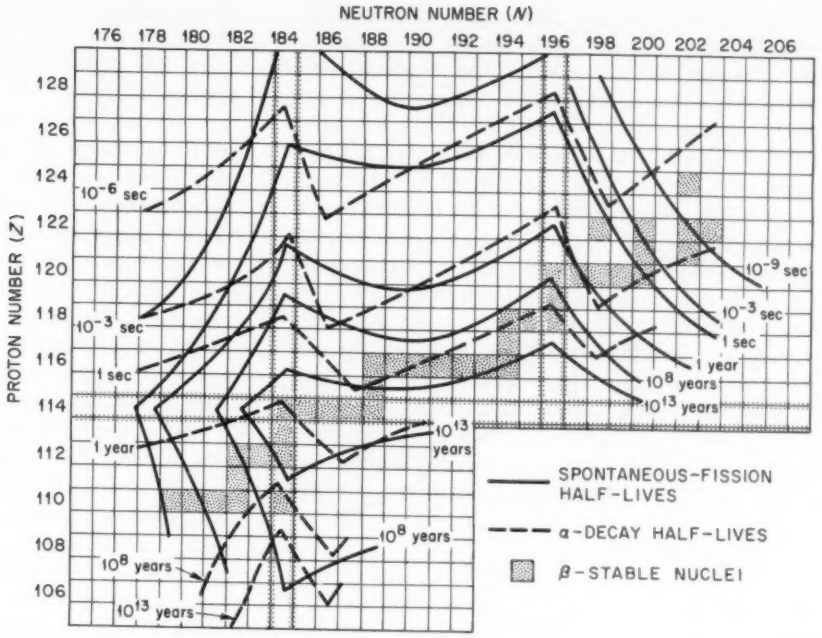

Fig. 4 Half-life contours for region of exceptional nuclear stability.

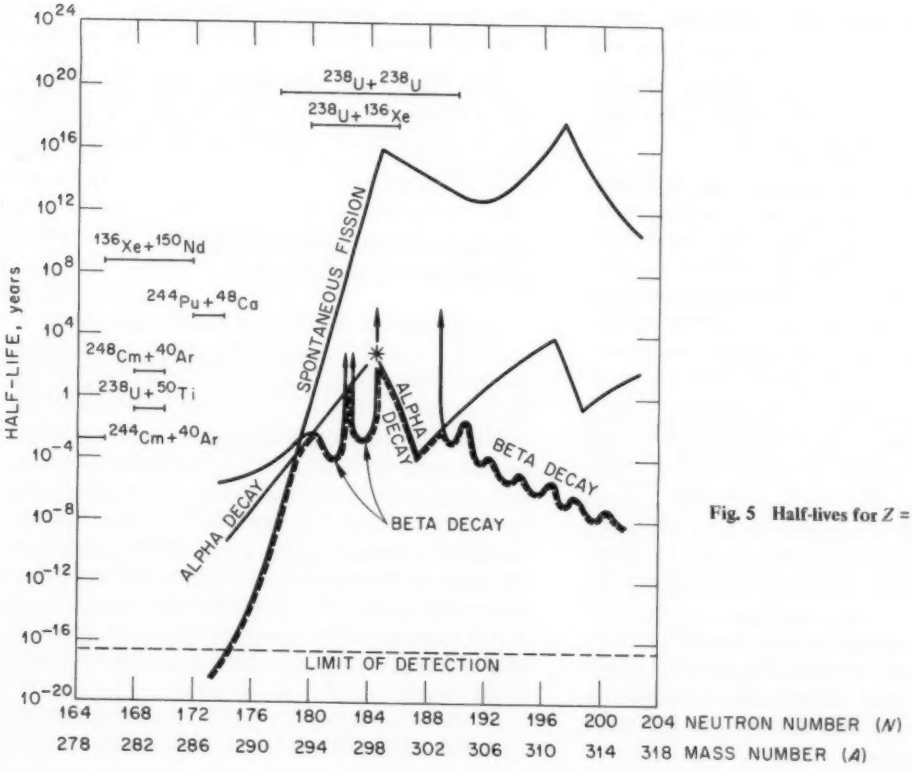

Fig. 5 Half-lives for $Z = 114$.

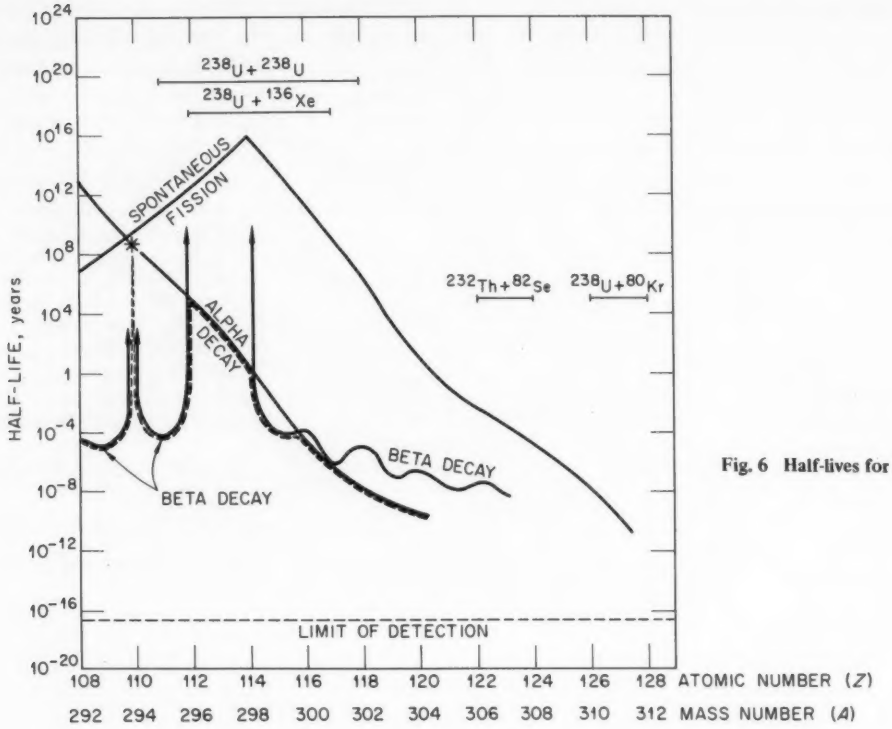

Fig. 6 Half-lives for $N = 184$.

taking into account data from Nilsson, Tsang, Swiatecki, and others, and using some of my own estimates for alpha and beta stability.[12b]

In Fig. 5, half-life values for spontaneous fission and for alpha- and beta-particle decay are plotted as functions of neutron number. The dotted line depicts the net or effective half-life for isotopes of element 114, since it follows the controlling mode of decay (the lowest of the three lines representing the three modes of decay). It can be seen that maximum stability, i.e., a half-life of about 10^3 years, occurs at $N = 184$, which corresponds to the isotope $^{298}114$. However, a large region of neutron numbers (or mass numbers) lies above the limit-of-detection line, which is set at 1 nsec (3×10^{-17} year). Note that $^{298}114$ should be beta stable, should have a negligible spontaneous-fission decay rate, and its rate of decay should be determined by alpha-particle emission with the half-life shown.

By holding the neutron number constant at 184 and varying the atomic number, Fig. 6 results. This figure demonstrates that theoretically it should be possible to observe many new elements with half-lives above the limit of detection.

The bars labeled with nuclear reactions in Figs. 5 and 6 represent the types of nuclear reactions available for the production of superheavy isotopes, and I shall have more to say about this later.

SEARCH FOR SUPERHEAVY ELEMENTS IN NATURE

It appears that maximum stability should be found at $Z = 110$ and $N = 184$ ($^{294}110$), an isotope for which the half-life might be as long as 10^8 years for decay by both spontaneous fission and alpha-particle emission and which is possibly stable toward decay by beta-particle emission. A half-life of a few times 10^8 years is sufficiently long to allow an isotope to survive and still be present on earth (for example, ^{235}U has a half-life of 7×10^8 years), provided it was initially present as the result of the cosmic nuclear reactions that led to the creation of the solar system. Referral to Figs. 1 and 2 will remind us that element 110 is a homolog of platinum and should have chemical properties similar to that precious metal.

Therefore searches have been made for element 110 (and neighboring elements) in naturally occurring

platinum and other metals and ores by workers at the Lawrence Radiation Laboratory in Berkeley and Livermore, at Argonne National Laboratory, at Oak Ridge National Laboratory, and at the Dubna Laboratory. The efforts of S. G. Thompson and associates at Berkeley are perhaps typical in this regard, and a brief description of their work will serve as an example of the complexities involved. They have looked for all types of radioactive decay; in addition, on the unlikely assumption that such elements might be so stable as to exhibit no detectable radioactivity, they have also employed very sensitive methods of nuclear analysis, such as X-ray fluorescence, mass spectrometry, and all types of activation analysis in their search. So far they have found no indications of the existence of such elements in nature and tentatively conclude that, if present, the concentration must be less than one part per 10 billion in platinum found in placer ores.

Flerov and coworkers at the Dubna Laboratory feel that they have detected spontaneous fission in lead samples and suggest that it might possibly be due to eka-lead (element 114); this would be inconsistent with the predictions summarized in Fig. 5, which indicate that the half-lives of the isotopes of element 114 are too short to allow their continued existence in nature today. Of course, the results of Flerov et al., if confirmed, could prove that the half-lives for alpha-particle decay have been underestimated. On the basis that the alpha half-lives may be substantially longer than indicated, consideration is being given to searches in nature for elements 111 (eka-gold), 112 (eka-mercury), and especially 113 (eka-thallium). Also, because elements with half-lives as short as 10^5 years could be present in cosmic rays, a search for superheavy elements is being made in this possible source.

METHODS FOR SYNTHESIS OF SUPERHEAVY ELEMENTS

From the foregoing account it is apparent that we now see a "promised land" of superheavy elements—a far-out island of nuclear stability—and we must devise means to reach it. The indications are that it can be reached only by bombarding target nuclei with sufficiently energetic heavy-ion projectiles. But there are many experimental difficulties to be overcome. The yield of the desired product nuclei is predicted to be very small because the overwhelming proportion of the nuclear reactions leads to the fission reaction rather than to synthesis through amalgamation of the projectile and target nuclei. The presently available target nuclei and projectiles lead to neutron-deficient nuclei that lie outside the perimeters of the island of stability. Projectile ions that will soon be available with the required energy should lead to nuclei just barely within the island and therefore to nuclei with half-lives so short as to make them difficult to detect. To reach the center of the island of stability may require the construction of new accelerators or the modification of present accelerators in order to furnish the unusual heavy ions that may be required at the necessary intensity and energy. The leading accelerators being devoted to this research are the HILAC at Berkeley and the cyclotrons at the Dubna Laboratory. These are already being modified to accommodate experiments of the type that will be needed to reach the island of stability.

The following is an example of a nuclear reaction that is relatively easy to carry out:

$$^{248}_{96}\mathrm{Cm} + ^{40}_{18}\mathrm{Ar} \rightarrow ^{284}114 + 4_0n^1$$

The mass number 284 shown is only illustrative, since a range of mass numbers can be expected as the number of neutrons emitted varies. The curium target and argon projectile at the required intensity and energy are available. The desired atomic number (Z) of 114 is achieved, but the neutron number (N) is only $284 - 114 = 170$, apparently placing the nucleus in the sea of spontaneous fission and making it unobservable. This is demonstrated graphically in Fig. 5. This reaction has been attempted at Berkeley, but no identifiable products have been formed.

Therefore a nucleus with additional neutrons is required to place it even at the edge of the island of stability. About the best that might be done in the near future is the reaction

$$^{244}_{94}\mathrm{Pu} + ^{48}_{20}\mathrm{Ca} \rightarrow ^{288}114 + 4_0n^1$$

producing a nucleus with 174 neutrons which is at the edge of the predicted island of stability (Fig. 5). This reaction uses ingredients that are relatively rich in neutrons. The isotope ^{244}Pu is a relatively rare material, but it will soon be available in sufficient quantity as the result of intensive neutron irradiation of ^{239}Pu and its transmutation products. The projectile isotope, ^{48}Ca, presents other problems; it occurs in nature only to the extent of less than 0.2% of the calcium isotopes. Therefore it must be enriched by mass-spectrographic techniques to a much higher proportion if it is to be an effective reactant. And it is difficult to vaporize and ionize so that it can be accelerated, although new

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devices for accomplishing this are being designed and tested. Other neutron-rich isotopes of interest as projectiles include ^{64}Ni and ^{50}Ti (Fig. 5), but they present some of the same problems as ^{48}Ca.*

Even reactions with heavier ions, of a type where no neutrons are emitted, such as

$$^{150}_{60}\mathrm{Nd} + ^{136}_{54}\mathrm{Xe} \rightarrow ^{286}114$$

produce nuclei which are deficient in neutrons and therefore do not provide an entree into the island of stability (Fig. 5). More likely, of course, is the production of isotopes of element 114 with even lower mass numbers, corresponding to the emission of neutrons in the synthesis reaction. However, acceleration of ions as heavy as xenon, in quantity, is not possible in existing accelerators. It is possible to write reactions with such intermediate heavy ions which suggest the production of isotopes such as the desired ^{298}114, but they are of a type not expected to occur to any appreciable extent.

Perhaps the only way to provide the required ratio of neutrons to protons will be to use as ingredients nuclei like the very neutron-rich ^{238}U nuclei in reactions such as

$$^{238}_{92}\mathrm{U} + ^{136}_{54}\mathrm{Xe} \rightarrow [^{374}146] \rightarrow ^{298}114 + ^{72}_{32}\mathrm{Ge} + 4_0n^1$$

or

$$^{238}_{92}\mathrm{U} + ^{238}_{92}\mathrm{U} \rightarrow [^{476}184] \rightarrow ^{298}114 + ^{170}_{70}\mathrm{Yb} + 8_0n^1$$

Probably the best prospect is the second reaction because this makes the most neutrons available. The theory here is that if an unstable, excited nucleus—and maybe "nucleus" is not even a good descriptive term in this case—could be produced by the interaction or "fusion" of a uranium ion with uranium (or by a xenon ion with uranium), it would promptly decay by "fission" to products that were relatively stable. Actually, what is a more probable course of events would be the direct transfer of nucleons between the two reacting nuclei, with the product nuclei being formed directly and with no intermediate heavy nucleus being formed at all. Of course, a whole spectrum of products would be formed, but among them might be the desired superheavy elements. To accomplish the U–U reaction, uranium ions accelerated to about 1.5×10^9 ev would be required. Each ion must be multiply charged to a high degree by the removal of orbital electrons in order to enable it to be accelerated in a machine of reasonable size. No machines are presently available, either in the United States or the Soviet Union, which are capable of accelerating such heavy ions to energies sufficient to overcome the coulomb repulsion barrier. These machines can be built or present machines can be modified, however, with essentially no limit on the atomic number of the accelerated ions.

Figure 5, which summarizes where we are and where we hope to be, shows bars superimposed to depict the product isotopes that might be obtained from the various nuclear reactions that I have just discussed. The bars apply only to the horizontal scale; their location on the vertical axis is arbitrary and has no significance. It can be seen that nuclear reactions which are now (or soon will be) achievable fail to reach the region of interest because insufficient numbers of neutrons reside in the product nuclei. Only the ^{238}U–^{238}U reaction (or possibly the ^{238}U–^{136}Xe reaction) provides enough neutrons to reach the center of the island of stability.

Likewise, in Fig. 6, nuclear reactions such as

$$^{232}_{90}\mathrm{Th} + ^{82}_{34}\mathrm{Se} \rightarrow ^{307}123 + {}_1p^1 + 6_0n^1$$

and

$$^{238}_{92}\mathrm{U} + ^{80}_{36}\mathrm{Kr} \rightarrow ^{311}127 + {}_1p^1 + 6_0n^1$$

can be postulated which have the required neutron number of 184 in the product isotopes, but the number of protons (i.e., the atomic number) is too large for the region of maximum stability. Again, the U–U or U–Xe type of reactions appears most suitable.

If the superheavy elements can be produced and are as stable as the predictions indicate, proof of their existence will require the use of chemical as well as nuclear detection techniques. The chemical separations must be designed on the basis of some guidance as to the nature of the element sought, and thus predictions of the electronic structure and chemistry of the new elements may well be imperative. (PSB)

*In the enriched-stable-isotope inventories of the Isotopes Division at ORNL, there are gram quantities of these three isotopes enriched as follows: ^{48}Ca, >95%; ^{50}Ti, >80%; ^{64}Ni, >97%.— *The Editor.*

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Section

II

Isotopes and Radiation Technology

Isotope Technology Development

Low-Energy Radioactivity and Avalanche Semiconductor Radiation Detectors*

By P. A. Johnston, G. C. Huth, and R. J. Lockert

Abstract: *The technology of avalanche semiconductor radiation detectors is reviewed briefly, and the advantages of avalanche detectors are illustrated by a discussion of various applications, including X-ray fluorescence measurements, beta-particle detection, and tracer usage—especially in medicine. The chief advantages claimed for this type of detector are that it is windowless, has high internal gain, is rugged in construction, and is able to operate satisfactorily over a wide range of temperatures.*

Avalanche radiation detectors have been steadily developed at General Electric Company (GE) under the sponsorship of the Atomic Energy Commission since 1962. The program was initiated by the Division of Biology and Medicine (DBM) to capitalize on the basic usefulness of the surface contouring technique to produce a practical internally amplifying semiconductor radiation detector. The Division of Isotopes Development (DID) has further supported the nonmedical applications of avalanche detectors for use where environmental extremes or restrictions of size and fragility prohibit the use of other nuclear radiation detectors.

The present article describes briefly avalanche detector technology and discusses some of the applications being investigated at this time under joint DBM–DID support.

*Based on a paper presented at the Third Isotopes Applications Conference, Gatlinburg, Tenn., Apr. 27–30, 1969.

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DETECTOR TECHNOLOGY

The avalanche diode is the solid-state analog of the gas proportional counter. A typical example of proportional action in an avalanche diode is shown in Fig. 1, together with a comparable spectrum produced by a gas (argon) proportional counter. Although these spectrums compare well, a larger number of low-energy events probably occur in the avalanche diode because of the nonuniformity of the gain across the surface of the crystal. An example of gain nonuniformity is shown in Fig. 2, which is the gain pattern of a diode measured with a flying spot scanner. The nonuniformity of gain results from the difference in degree of resistivity uniformity available in the basic semiconductor crystal, which is the determining factor in establishing the magnitude of the electric field in the crystal.[1] The attainment of uniform resistivity continues to be the major technological problem that must be solved before the avalanche detector will be useful in nuclear spectroscopy.

Most of the advantages in the use of the avalanche diode as a particle detector accrue as a result of the high speed (less than 2×10^{-8} sec) of the multiplication process. Thus, since the energy of nuclear particles is transformed in about 10^{-12} sec and noise electrons drift into the junction in about 10^{-6} sec, then if the charge can be collected in less than 10^{-8} sec (as the avalanche diode does), a high ratio of signal-to-noise electrons can be realized. The tunnel diode is an excellent electronic circuit element for making the required detection decision in the nanosecond time domain, and the combination of the avalanche diode and the tunnel

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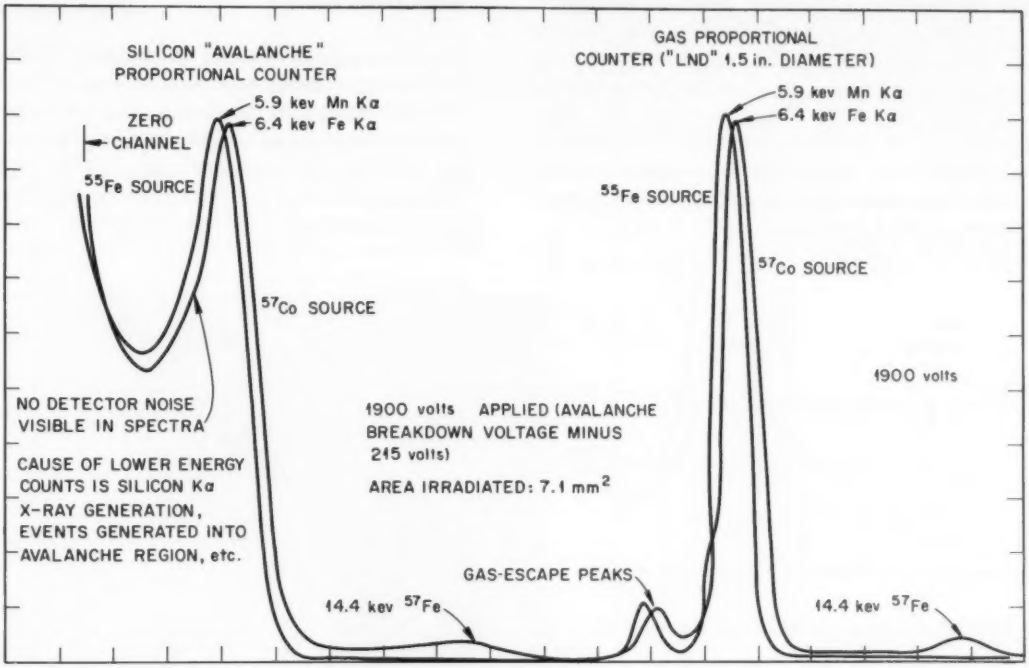
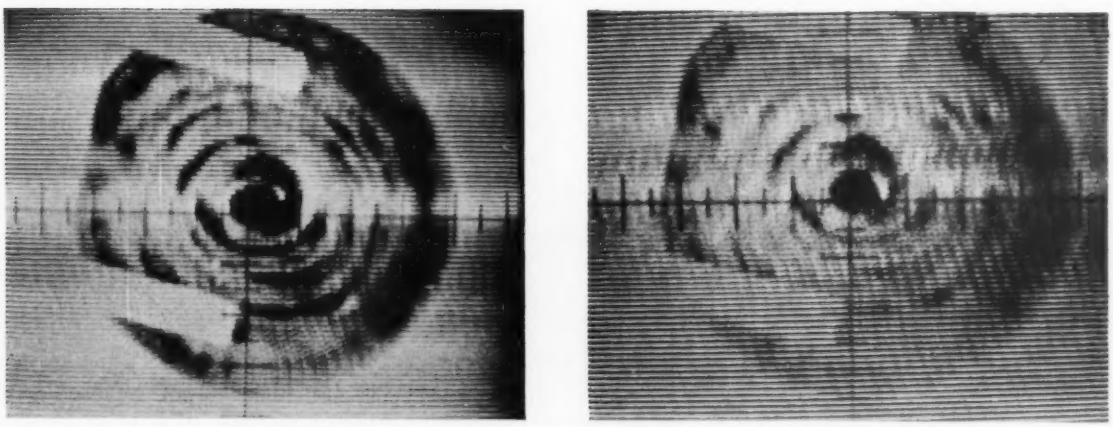

SILICON "AVALANCHE" PROPORTIONAL COUNTER
ZERO CHANNEL
5.9 kev Mn Kα
6.4 kev Fe Kα
55Fe SOURCE
57Co SOURCE
NO DETECTOR NOISE VISIBLE IN SPECTRA
CAUSE OF LOWER ENERGY COUNTS IS SILICON Kα X-RAY GENERATION, EVENTS GENERATED INTO AVALANCHE REGION, etc.
1900 volts APPLIED (AVALANCHE BREAKDOWN VOLTAGE MINUS 215 volts)
AREA IRRADIATED: 7.1 mm2
14.4 kev 57Fe
GAS-ESCAPE PEAKS
GAS PROPORTIONAL COUNTER ("LND" 1.5 in. DIAMETER)
5.9 kev Mn Kα
6.4 kev Fe Kα
55Fe SOURCE
57Co SOURCE
1900 volts
14.4 kev 57Fe

Fig. 1 ^{55}Fe and ^{57}Co X ray.



(a) MODERATE GAIN SHOWING EXTENT OF CORE

(b) HIGHER GAIN YET IMMEDIATELY BEFORE AVALANCHE BREAKDOWN

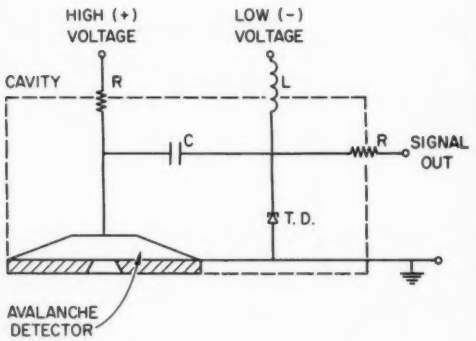
NOTE: HIGH GAIN INDICATED BY DARKER AREAS IN THESE SCANS

Fig. 2 Result of scanning P+ side of entire wafer (~2.5 cm in diameter) of crystal grown by one manufacturer, showing coring (*a*) and fine macrostriations (*b*).

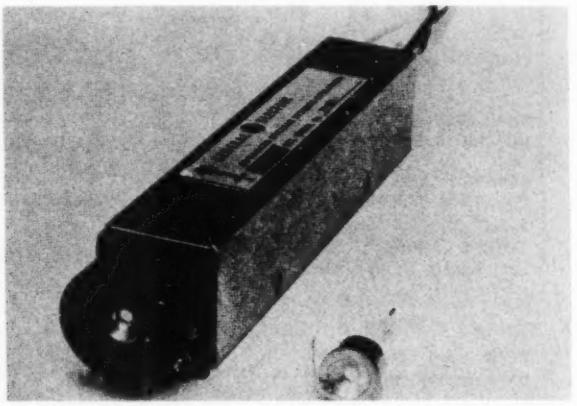
diode discriminator is called the "avalanche detector."[2] An example of the kind of sensitivity that this fast time technique makes possible is the detection of the aluminum fluorescence X ray (1.49 kev) at a detector temperature of 100°C. Figure 3 shows the basic schematic of the avalanche detector. The temperature-compensation aspect will be discussed, together with the high-temperature counting-efficiency measurements, in the following section.

APPLICATIONS

The avalanche detector is essentially windowless and can thus detect any nuclear radiation from the ultrasoft X ray to minimum ionizing events. In practical applications, radiations should be used that are absorbed in the drift region (Fig. 4) of the avalanche diode because multiplication decreases with increasing penetration into the crystal.



(a) SCHEMATIC DIAGRAM



(b) MARK III DETECTOR ASSEMBLY WITH SOLID-STATE TEMPERATURE CONTROLLED HEATER

Fig. 3 Avalanche detector–tunnel diode circuit and sample device.

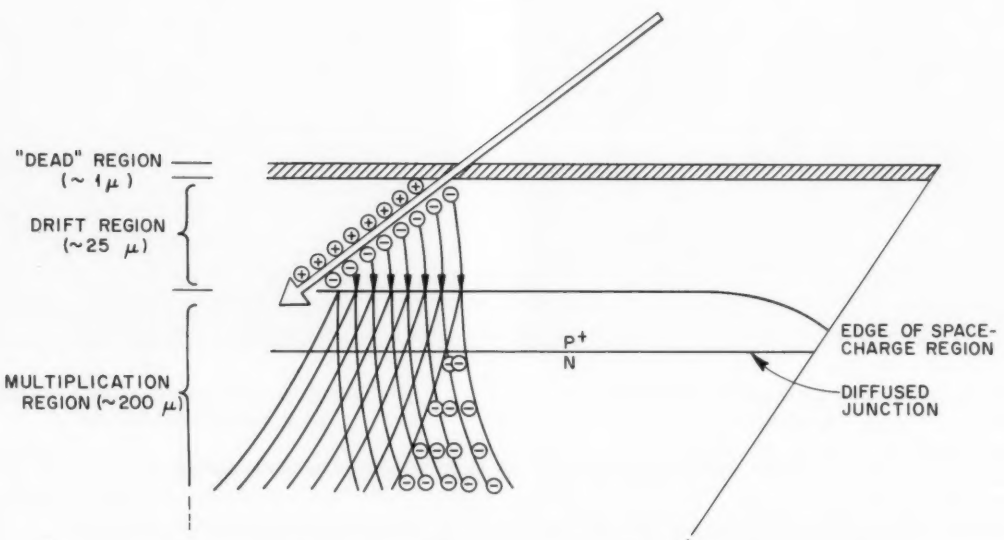


Fig. 4 Operation of internally amplifying detector.

X-Ray Applications

Although the avalanche detector has had some value in the past in X-ray astronomy, most recent applications have been in X-ray fluorescence (XRF). For example, last year DID initiated a collaboration between workers at General Electric (GE) and Trombka and Adler of NASA Goddard Space Flight Center to determine if an X-ray fluorescence instrument could be developed for planetary geochemical exploration. Figure 5 shows an artist's sketch of the

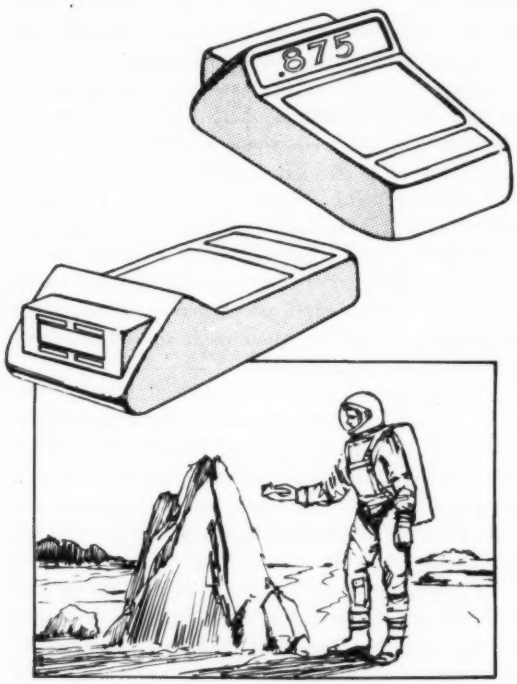


Fig. 5 Artist's concept of hand-held analyzer.

concept, i.e., a hand-held analyzer that could be used to help astronauts select lunar rock specimens to be returned to earth for laboratory analysis. Avalanche detectors are well suited to applications of a geological nature because the characteristic X rays emitted by the common rock-forming elements (Mg, Al, Si, Ca, K, and Fe) correspond with the spectral response of the detector shown in Fig. 6. Also, as is typical of solid-state equipment, avalanche detector instrumentation can readily be made portable for field applications.

The X-ray energy discrimination in prototype instruments developed for this application is provided by "balanced filters"—a technique perfected by John Rhodes of Columbia Scientific Research Institute. Our concept, as shown in Fig. 7, uses two detectors: one

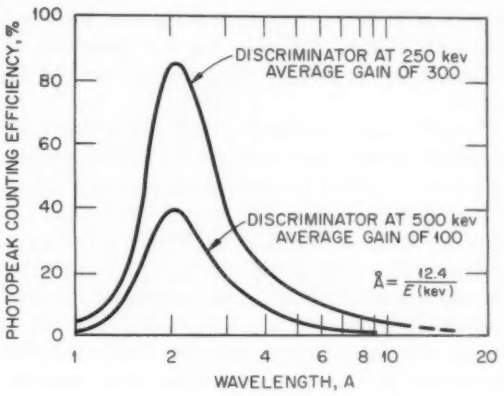


Fig. 6 Spectral response of silicon, avalanche radiation detectors.

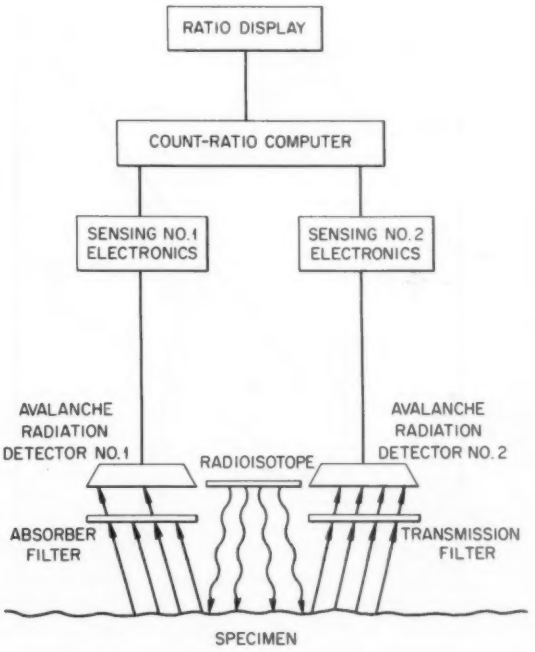


Fig. 7 Principle of operation of the General Electric XRF analyzer.

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with a transmission filter, and the second with an absorber filter. The events counted in each filtered detector are electronically subtracted in real time so that the remaining events are due only to X rays that have energies between the absorption edges of the two filters. This real-time aspect, as opposed to mechanically positioning filters in front of a single detector, is made possible because the detector is very small and thus both filtered detectors can be placed in an efficient geometrical arrangement of the source and the detector.

Silicate rock standards supplied by the U. S. Geological Survey were analyzed for their potassium and calcium contents by a laboratory prototype of the instrument that incorporated a 10-mc ^{55}Fe source. The results are shown in Fig. 8. In addition, a soil-analysis experiment for a Mars lander has been generated from this collaboration. The designed equipment will use a single avalanche detector and a series of Ross filters to analyze, with the aid of an earth-based computer, the fluorescence X rays produced when alpha particles from a radioisotope source strike the Martian surface. The principal advantage of this instrument is a 3-min analysis for the six common rock-forming elements as compared to several hours' counting time required for the alpha-backscatter analyzer on Surveyor.

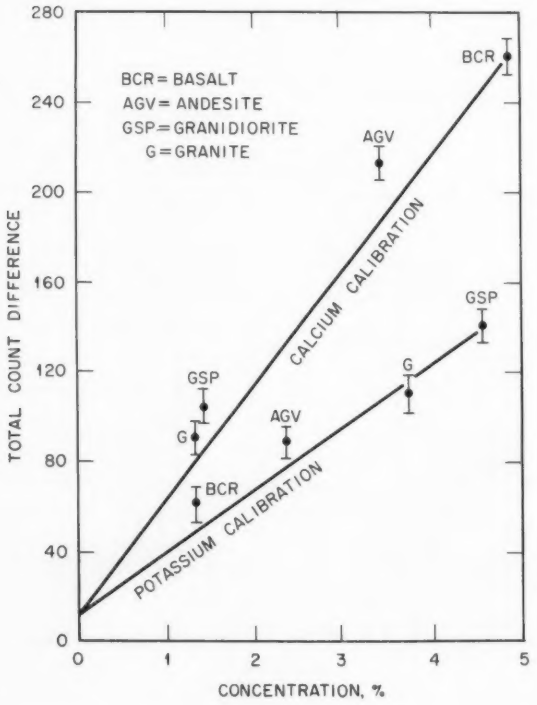

Fig. 8 Analysis of calcium and potassium contents.

Most of the present X-ray fluorescence work is aimed toward developing a hand-held, small area, X-ray fluorescence probe for use in dental and geological applications. The dental work is being carried on through a collaboration with Dr. Robert Besik of the University of Chicago. The purpose of this effort is to investigate the feasibility of performing in situ tooth-calcification studies with an avalanche detector XRF. The application is ideal for an ^{55}Fe radioisotope XRF analyzer, because human teeth contain negligible amounts of those elements that could interfere with the calcium determination.

In another application an attempt is being made to use the avalanche detector XRF probe to determine the potassium content of single mineral crystals on the surface of a rock. This capability in a portable instrument is being sought by the U. S. Geological Survey so that the cooling patterns of large rock formations can be identified in the field rather than by the usual time- and money-consuming methods of collecting, tagging, and transporting samples to a petrographic laboratory where either optical or microprobe analysis is performed.

For gamma-ray dosimetry, silicon avalanche detector performance was assessed by Jones[3] working at the Atomic Energy of Canada's Chalk River Nuclear Laboratories. From his experiments he concluded that this was suitable for measuring gamma exposures and dose rates over a wide range of gamma energies (30 kev to 1.25 Mev) and over a wide range of exposure rates (from background up to 1 r). His data on gamma sensitivity are shown in Fig. 9. He has since developed an interesting prototype of an area-monitoring instrument that is capable of simultaneously monitoring exposure rate and integrating the total exposure—a function that cannot be performed by either ion-chamber or Geiger-tube detectors.

Beta-Particle Applications

The avalanche detector is well suited to beta-particle applications because of high specific energy deposition in the crystal. In comparison with other devices equally suited for this purpose, the avalanche detector offers two advantages: it is windowless and it has internal gain. Since it has no window, the avalanche detector can detect very-low-energy beta particles; as shown in Fig. 10, the low-energy cutoff because of the dead layer occurs just below 5 kev. Present research is

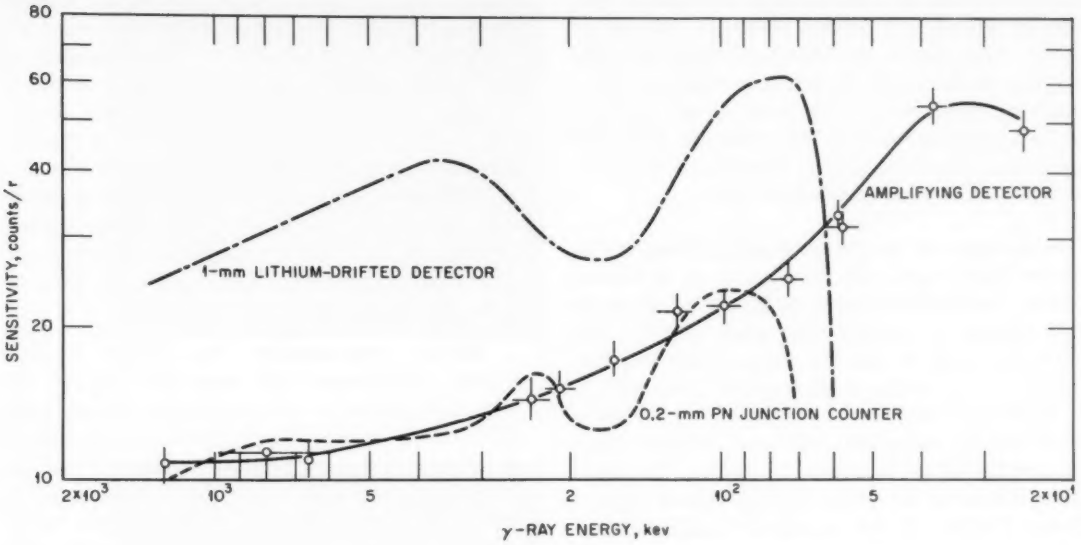

Fig. 9 Dependence of sensitivity on X-ray energy.

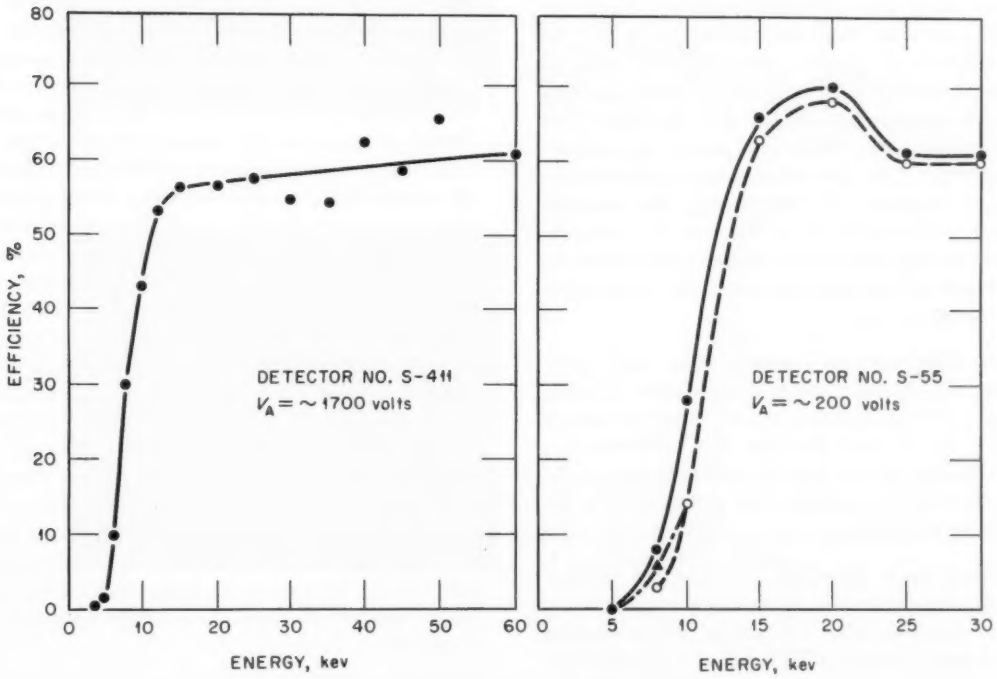

Fig. 10 Efficiency of detectors S-411 and S-55.

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aimed at lowering the cutoff energy even further. The effect of internal gain on beta detection is shown in Fig. 11. This shows the excellent signal-to-noise advantage of the avalanche detectors relative to non-amplifying silicon devices. Obviously, in any application, a high signal-to-noise ratio leads to improved performance. However, in space experiments, it can be the difference between success and failure.

Measurement of the Earth's Magnetosphere. In the last few years, work has been going on at NASA Goddard Space Flight Center under the direction of Dr. D. Williams to measure low-energy electrons in near space to aid in defining and understanding the dynamics of the earth's magnetosphere.[4] One part of the experiment has been the measurement of low-energy-electron intensities. Previously, this was done with surface-barrier detectors. In general, the best devices obtainable have noise levels of around 24 kev (10-kev FWHM). In the operational experiment, in order to ensure that noise generated background would not interfere with data during flight, the detection threshold is set at nearly twice the noise level. Thus the lowest energy electron detectable in this experiment is approximately 40 kev with the surface barrier. However, by using the avalanche detector, which has essentially the same noise level as a surface barrier, the same equipment is capable of detecting 5-kev electrons because of internal gain, since the avalanche detector having an average gain of 200 will multiply 5-kev events out to 1 Mev. Thus, with such a large signal-to-noise ratio (25), low-energy-electron detection is no longer marginal. At this writing, the expected improved performance from the use of avalanche detectors is being verified on sounding rocket flights by Williams and his colleagues prior to earth orbital flights on the IMP spacecraft.

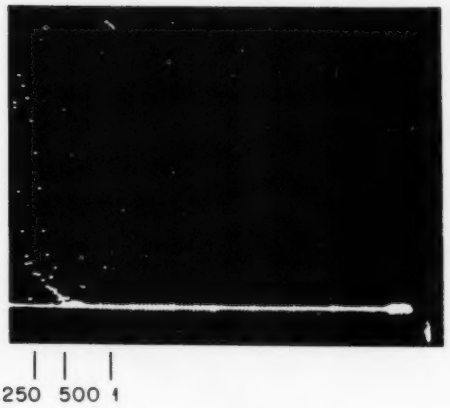
Mars Life-Detection System. The Mars life-detection experiment requires a sterilizable detector that has a low-background, high-counting-efficiency capability for ^{14}C beta particles. With avalanche detectors, the background rate for 50% detection of ^{14}C is approximately 4 counts/hr and performance is not degraded by sterilization.

Thruster Leak Detection. An in-flight thruster leak-detection system is being developed, in conjunction with the Jet Propulsion Laboratory, which requires mixing a small amount of ^{85}Kr with the propellant gas and measuring leaks as low as 0.001 cm^3/hr by detecting the 667-kev beta particles from the leaking ^{85}Kr. Again, leak sensitivity is obtained because low background and high detection efficiency are provided from the internal gain of the avalanche detector. For space applications in general, the relative simplicity of this detection system promises a high resistance to launch vibration and high g loadings from impact. Power, volume, and weight requirements are relatively low. Again, of special significance for extraterrestrial applications is that performance of the avalanche detector should not be affected by exposure to a heat sterilization cycle. Such a cycle has resulted in degraded performance of the gas-type nuclear detectors in the past.[5]

Density Measurements. The simple, accurate density measurement has long been one of the principal applications of radioactivity, and avalanche detectors are not only broadening the sensitive range of these measurements (down to 10^{-6} g/cm^3) but are allowing density measurements to be performed in extremely hostile environments. For example, Industrial Nucleonics (Columbus, Ohio) has recently considered avalanche detectors for use in the inlet shock-position sensing system of the supersonic transport (SST)[6] shown in Fig. 12. If the shock wave is too far into the throat of the jet engine on the SST, the efficiency of the system is reduced, which in turn increases cost through excessive fuel consumption.

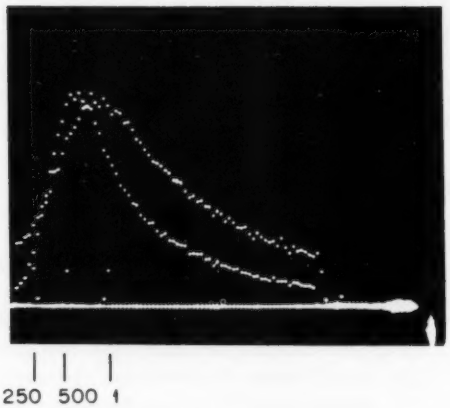
In this application, conventional pressure-cell measurements cannot be used to control shock-wave position because they introduce inaccuracies due to lambda shock waves and boundary-layer effects. The radioisotope density gage can provide better control of the normal shock position because it is not subject to these inaccuracies. By the use of avalanche detectors, it should be possible to obtain maximum sensitivity to shock position because a low-energy beta particle from ^{85}Kr or ^{14}C can be used without regard for bremsstrahlung interference. More significantly, the counting efficiency can be kept relatively constant in such a harsh thermal environment by surrounding the detector with a small oven, shown previously in Fig. 3. This kind of heat stabilization at high temperature is unique to the avalanche detector, since all other nuclear detectors must be cooled in hot environments before the thermally generated noise increases to the point where it swamps the signal. On the other hand, the avalanche detector can be operated at temperatures in excess of 125°C without measurably reducing the signal-to-noise ratio.[7]

Measurement of Shock-Front Magnitude. In a similar application, work has begun at the Suffield Experimental Station in Ralston, Alberta, Canada, to

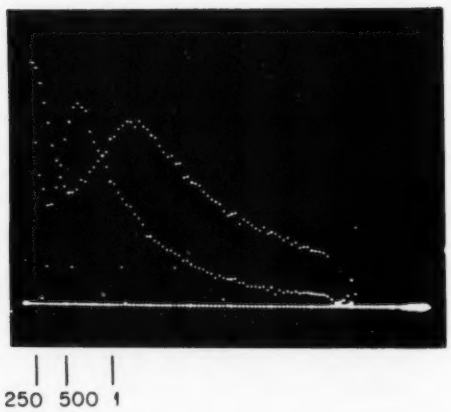


^{106}Ru AND ^{85}Kr DETECTION COMPARISON

ORTEC SURFACE BARRIER
50 volts
MARKERS AT 250,500 kev AND 1 Mev
LEFT: ^{106}Ru RIGHT: ^{85}Kr



LND PROPORTIONAL COUNTER
1950 volts
2-mil Be WINDOW
MARKERS AT 250,500 kev AND 1 Mev
LEFT: ^{106}Ru RIGHT: ^{85}Kr



AVALANCHE DETECTOR
1800 volts
MARKERS AT 250,500 kev AND 1 Mev
LEFT: ^{106}Ru RIGHT: ^{85}Kr

Fig. 11 Effect of internal gain on beta detection.

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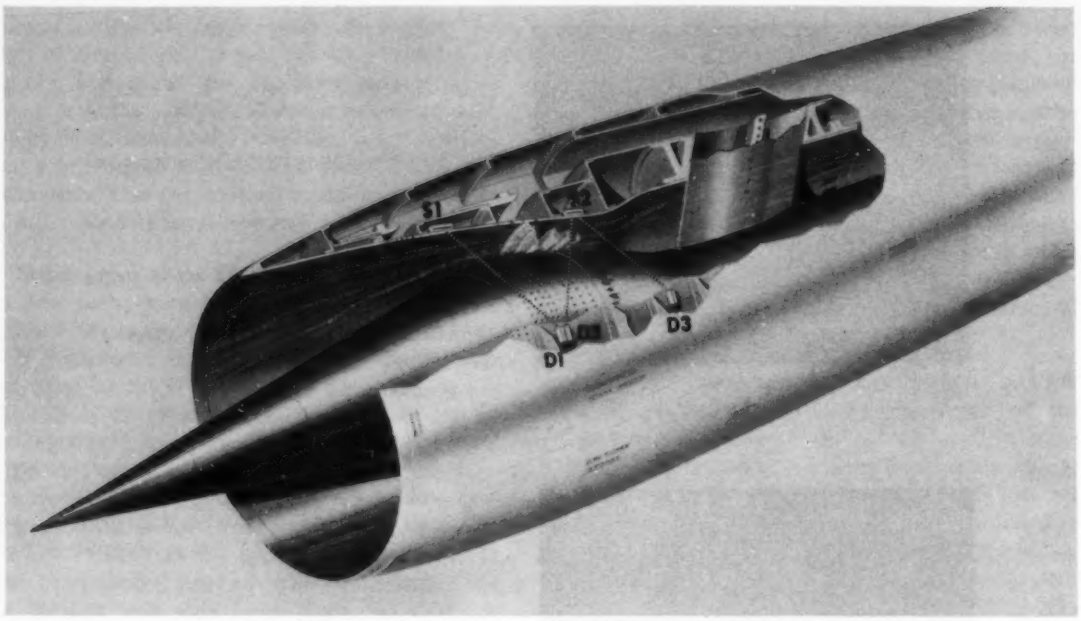

Fig. 12 Conceptual design of a nucleonic inlet control system. (S = sources; D = detectors.)

measure, with an avalanche detector and a ^{147}Pm source, the magnitude of a shock front as it travels down a shock tube following a 2-ton TNT blast. In this use, advantage is taken of the high-count-rate capability of the detector. Since the density change must be measured in about 1 msec, the avalanche detector must count the beta particles at a rate of 10^6/sec in order to obtain accurate statistics. As shown in Fig. 13, this is well within the capability of the detector.

Tracer Applications

Most tracer applications involving avalanche detectors have been in the field of medicine; however, because of the low background and high detection efficiency for low-energy beta particles, an effort has been made to determine the feasibility of using the detector as a solid-state tritium counter.

The entrance dead layer of the detector is approximately 1 μ of silicon, which represents the depth of penetration of a 10-kev beta particle; thus one can expect that detection efficiency for ^3H will be low. In spite of this limitation, the promise of a portable tritium counter for field use in such areas as hydrology and agriculture seems to make the investigation worthwhile. Subsequent measurements[8] carried out in 1967 by V. Gelezunas of the GE laboratory on a device having a 500-kev tunnel diode discriminator resulted in a tritium counting efficiency of 0.001. However, with the lower energy discriminator of present detectors, the detection efficiency may be as high as 1%. Tritium counting efficiency then is low; however, it is more than sufficient for precise measurements because the background is less than 1 count/min. With such a background the minimum sensitivity for the portable instrument should be around 10^{-11} curie—an order of magnitude more sensitive than Geiger-counter systems. It is interesting to note that, for ^{14}C tracing, the minimum sensitivity is around 10^{-13} curie because counting efficiency (50%) can be sacrificed to obtain very low backgrounds on the order of 4 counts/hr.

Medical Applications

At present, avalanche detectors are being evaluated for use in a number of medical applications. Two applications that best exemplify the medical advantages obtained by using these detectors are discussed.

Heart Leakage Diagnosis. A collaboration was initiated through DBM with Dr. Dean Mason of Cardiovascular Diagnosis, Cardiology Branch, National

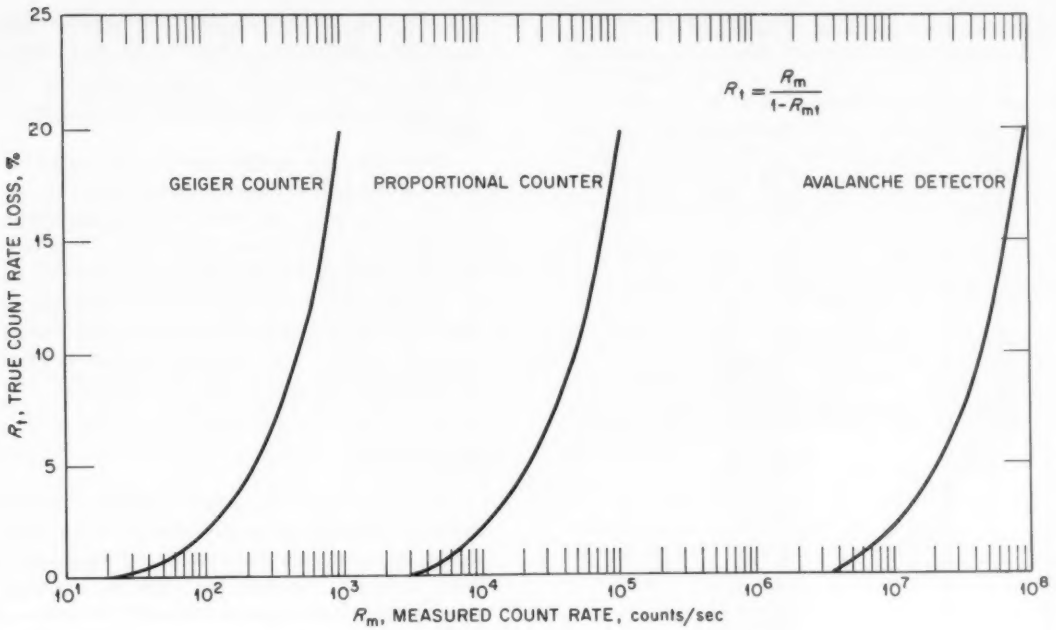

Fig. 13 Theoretical count-rate capacity of some nuclear detectors.

Heart Institute. This work has involved the development of an avalanche detector catheter for use with Mason's technique for diagnosing one of the most common types of congenital heart disease–i.e., intracardiac defects or holes in the dividing wall (septum) between the right and left chambers of the heart.

As originally developed, his technique required the patient to inhale ^{85}Kr into the lungs and thence to the left side of the heart. The appearance and the amount of ^{85}Kr in the right side of the heart was then determined by removing blood samples from the right chamber and counting externally with a Geiger counter. According to Mason, the ability to sense the radioactivity within the heart would first allow even small defects to be diagnosed but, more important, it would increase the investigator's ability to quantify the amount of blood that flowed through the hole. It is the amount of flow across the defect that forms the basis for determining whether or not an operation is necessary to close the defect. The ^{85}Kr catheter detector delivered to the AEC is shown in Fig. 14. The mounted detector (on a 36-in.-long catheter) has a 200-volt avalanche voltage, and ^{85}Kr detection with it has been found to be relatively insensitive to small temperature changes around 37°C—a significant attribute for body implantation.

The development of the catheter detector points the way to improved performance for implantable detectors because avalanche gain can again be used to lower the present practical detection threshold set by thermal noise generation, from a 300-kev beta particle to a 10-kev beta particle at body temperature. The advantages gained are the choice from among many low-energy-beta-emitting isotopes and the detection of beta particles whose ultimate energy will be very low because of scattering by the body tissues.

Mapping of Wound Sites. Aside from the advantages an internal gain device offers in extralaboratory environments, the small size of the avalanche detector makes it of value in many medical applications. Much of the work supported by DBM has been related to the problem of locating plutonium in wounds through the detection of the daughter-product 17-kev X-ray emission. The portable instrument developed for this purpose is shown in Fig. 15. The avalanche detector is housed at the end of the hand-held probe, while the necessary power supplies, batteries, and digital display are contained in the electronic counter. In recent experiments conducted in collaboration with Dr. Robert Waters at Colorado State University, the instrument was used to map the wound

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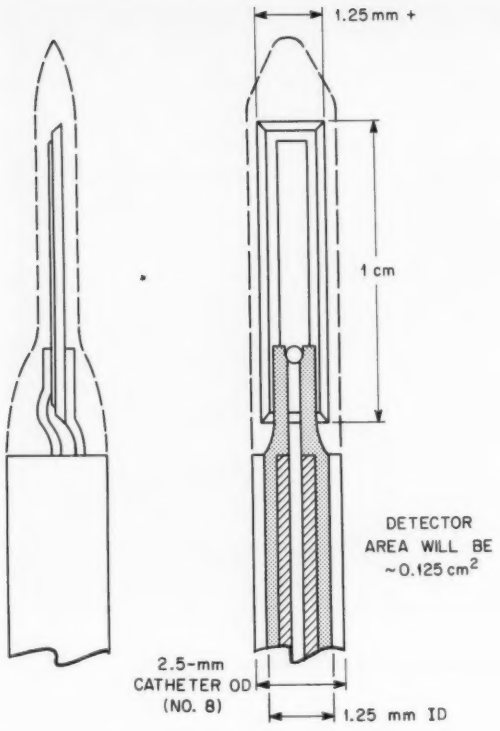

Fig. 14 Krypton-85 catheter detector.

site of a 2-μc, subdermal, plutonium injection given to an animal 5 days prior to the test. In comparison with a 1-in.-diameter scintillator, the wound could always be mapped much more precisely with the avalanche detector.

Obviously, the smaller size of the avalanche detector probe defines more accurately where the wound boundary occurs, but, more importantly, when the wound was scanned, it was much easier to tell when a boundary was reached because the change from signal to background (5 counts/sec to 0.002 count/sec) obtained with the avalanche detector was much more graphic than the change (60 counts/sec to 1.6 counts/sec) obtained with the scintillator.

SUMMARY

An effort has been made to illustrate the advantages of avalanche detectors through a discussion of various applications. Comparisons have been made with other detection systems but only as a means of showing that internal gain in avalanche detectors makes solid-state nuclear instrumentation practical because of its similarity in mode of action to that of the gas-filled Geiger tube. Certainly the improving skill in the use of avalanche detection will bring to reality those applications presently considered feasible, but the hoped-for end result of research will be the

Fig. 15 Portable avalanche detector instrument.

discovery of new applications that require a very small detector sensitive to low-energy radioactivity and capable of yielding higher count rates than have ever been achieved outside of special laboratory environments. (FJM)

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Evaluation of Cracks in Turbine-Blade Leading Edge by Use of ^{85}Kr Gas*

By William C. Eddy, Jr.†

Abstract: *Turbine blades were examined by use of radioactive ^{85}Kr gas as a tracer to locate otherwise undetectable cracks. The work reported includes that performed under a U. S. Air Force (USAF) contract and an in-house funded project in which blades were treated by KET.‡ The residual activity found 1 day after ^{85}Kr impregnation by use of KET was about $^{1}/_{50}$th that of the initial activity. A detection system that incorporates fast-response electronics provided excellent spatial resolution of the detected cracks in less than 1 min at those activity levels remaining 3 hr after impregnation by KET.*

The purpose of the work carried out under a U. S. Air Force contract was to determine the feasibility of using a chemically inert radioactive gas as a tracer to detect cracks in turbine blades. Routine examination of turbine blades by this method was the primary goal.

The San Antonio Air Matériel Area (SAAMA) Service Engineering Division, USAF,§ supplied used turbine blades for examination by the tracer-gas technique. In this process a layer of radioactive ^{85}Kr was deposited on the metallic surface of a blade. The amount of radioactivity along the leading edge was then estimated by autoradiography and by instrumental means so as to correlate activity concentration with crack locations along the edge. The radiometric method was then intensively investigated to obtain a combination of excellent spatial resolution and short analysis time requisite for a production technique.

*Industrial Nucleonics Corporation, Evaluation of Cracks in Turbine Blade Leading Edge by Use of Krypton Gas, Final Report, Columbus, Ohio, Jan. 28, 1969.

†Industrial Nucleonics Corporation, Columbus, Ohio 43202.

‡Krypton Exposure Technique, a service mark of the Industrial Nucleonics Corporation.

§Industrial Nucleonics acknowledges the material and consultive support of E. D. Walker of SAAMA who was the technical liaison between the USAF and Industrial Nucleonics. Samples used in the contract were furnished by the SAAMA Service Engineering Division of Kelly AFB, Texas.

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TECHNICAL BACKGROUND

Kryptonation* is the incorporation of ^{85}Kr, a radioisotope that primarily emits beta particles, into the lattice structure of a test material. Industrial Nucleonics has for some time used the process of diffusion of ^{85}Kr gas into a material; the kinetic energy of the gas at high temperature and pressure is taken up in the formation of quasi-stable radioactive compounds. The resultant test material, called a Kryptonate,† incorporates the activity within its host lattice and does not release it until the lattice is chemically or physically disturbed. Kryptonated turbine blades maintain a constant specific activity with time and do not release their activity unless heated to a temperature above any to which they have previously been subjected. Cracks in the leading edge of the same turbine blades could be located by autoradiography since crack location was indicated by an excess of ^{85}Kr as compared to that on the undamaged portions of the blade. The exposure time required for the autoradiograph test was 2 to 5 days.

The aim of the contract investigation was to shorten the testing time and to determine if the turbine blades could be made sufficiently radioactive to permit radiometric crack detection without retention of sufficient activity to constitute a health hazard.

The classical description of kryptonation as a diffusion process indicates that the activity per unit area should be exponentially proportional to the temperature and directly proportional to pressure and to the square root of diffusion time. The parameters are ranked in order of their significance to the specific activity of the test material.

EXPERIMENTAL PROCEDURE AND RESULTS

To fulfill the contract, Industrial Nucleonics studied several problems arising from the use of the kryptonation process in production processes concerned with the routine inspection of used turbine blades. Kryptonation tests were made to assess the possibility of detecting cracks in the leading edges of turbine blades by simple autoradiographic and radiometric (electronic) measurement.

Kryptonation Procedure

For the first kryptonation, six used turbine blades were cut into 1- by 0.375-in. segments. This particular size fit into existing kryptonation vessels, and particular segments were chosen to contain one or more definite cracks along the leading edge. Such a crack is visible in the photograph of a turbine blade in Fig. 1. The close-up shows one very wide crack. Three other cracks may be seen, but with difficulty, only from the other side of this blade section. The sections were kryptonated 3 hr at a pressure of 517 cm Hg and a temperature of 149°C. The activity of the resulting blade sections was estimated at 0.06 $\mu c/cm^2$ from measurements made with an end-window G–M counter using appropriate corrections for geometry, absorption, and backscatter differences from a standard source.

Negative Electronic Results

A $^{3}/_{32}$-in.-diameter avalanche detector system was used to scan the leading edge of one blade section. The curves of measured count rate vs. distance along the leading edge with distance increments of 50 mils between count-rate measurements are shown in Fig. 2. The detector was moved parallel to the leading edge as indicated in the sketch in Fig. 2. Curve 1 is a scan with minimum distance between the leading edge and the detector; curve 2 is a repeat of the latter with blade sides shielded to remove possible interference; curve 3 is a repeat of the first scan after mechanically polishing the leading edge in an effort to increase the ratio of activity in a crack to activity on the surface; curve 4 is a scan using a 10-mil lead sheet between the source and detector, with the lead having a $^{1}/_{32}$-in. hole in it for increased spatial resolution. The ends of the leading edge are indicated by E_1 and E_2 on the distance axis, and the location of cracks determined from visual observation is indicated by C_1 to C_6. Several negative results are apparent in all the curves. None of the measurement processes indicated definite crack detection that could be correlated with visible cracks. Also, the large count-rate increase near E_2 was not due to a crack but rather was determined from autoradiography to be caused by an edge irregularity formed in sectioning the sample from the original blade. The activity estimate of 0.06 $\mu c/cm^2$ was made by assum-

*In this report, kryptonation is used to describe a process in which diffusion of krypton gas into the crystal structure is significant. It usually requires temperatures appreciably higher than room temperature.

†The term Kryptonate, used to describe the process of incorporation of ^{85}Kr within the crystalline lattice of a material, is a proprietary term since the process was developed and patented by the firm of Parametrics, Inc., now known as Panametrics, Inc. Much of the developmental work to extend the use of the technique was funded by the Division of Isotopes Development, U. S. Atomic Energy Commission.

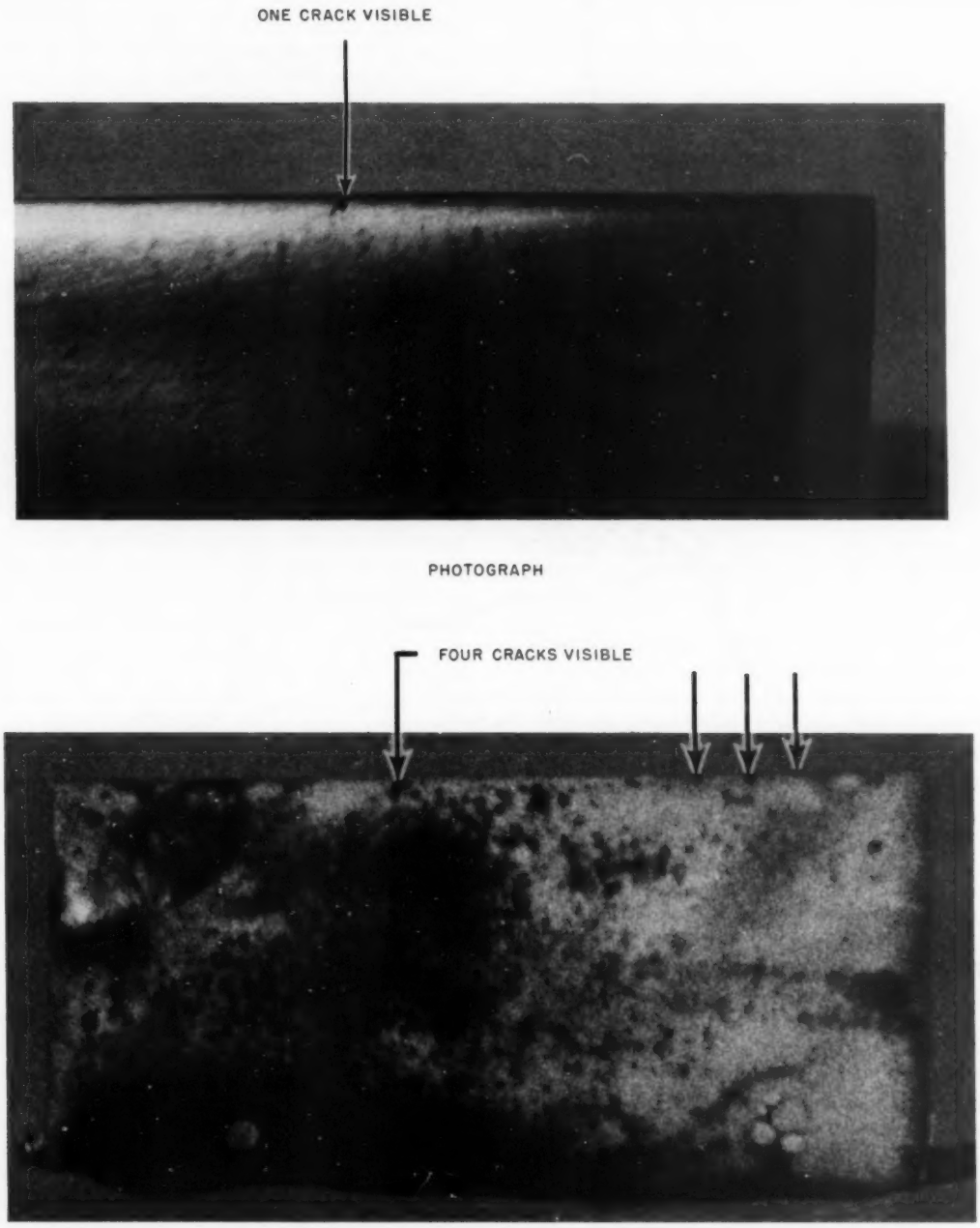

Fig. 1 Photograph and autoradiograph of test segments of turbine blade.

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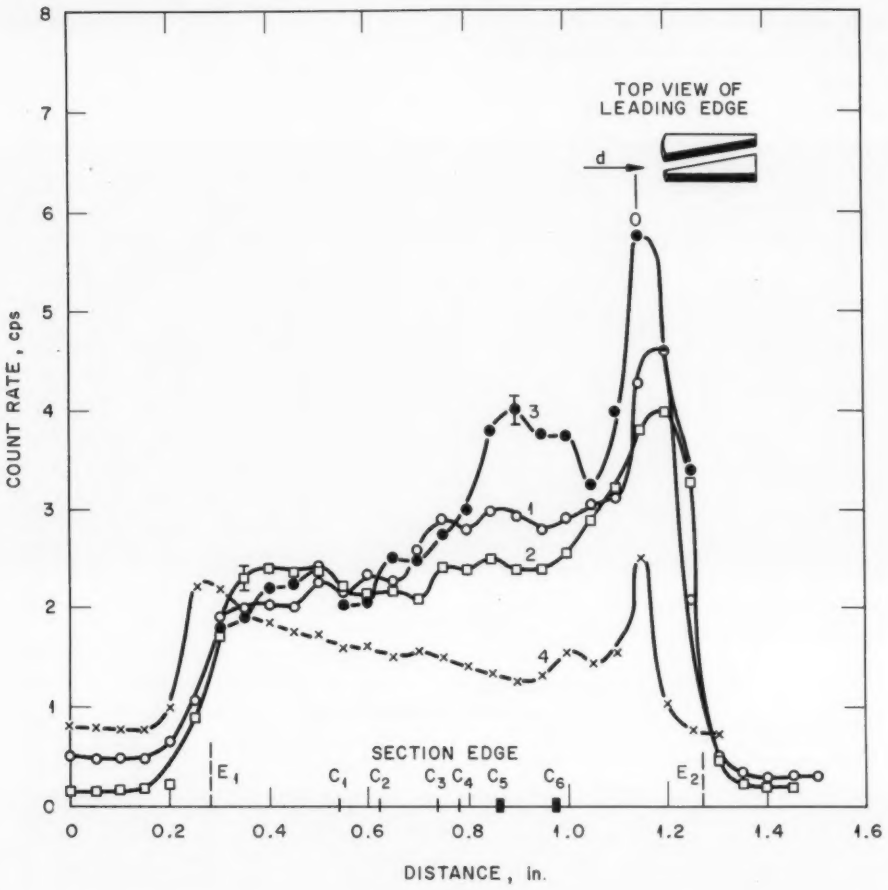

Fig. 2 Scans of turbine-blade sections. A 0.094-in.-diameter detector was used. Curve 1, minimum sample to detector distance scan. Curve 2, shielded sides, minimum distance scan. Curve 3, polished sides, minimum distance scan. Curve 4, 10-ml lead sheet with a 0.0312-in. hole, minimum distance, polished sides. Units on count-rate axis apply only to curve 3; all other curves are offset for clarity.

ing a uniform activity per unit area on a turbine blade; this procedure does not take into account activity concentrations at cracks. This activity implied a count rate of approximately 6 cps from the $\frac{3}{32}$-in.-diameter avalanche detector in the geometry used. The actual count rate varied from 2 to 6 cps along the leading edge. The results of these measurements were verified by using two other detection systems with both this blade section and another blade which showed definite cracks by autoradiography. A 1-in.-diameter silicon solid-state detector and a $\frac{1}{32}$- by $3\frac{1}{32}$-in. NaI(Tl) detector were used with a 10-mil-thick lead sheet, having a $\frac{1}{32}$- by $\frac{3}{32}$-in. slit in it, to make on- and off-crack measurements. In none of the three systems was it possible to detect a difference in counting measurements of <10% with the silicon detector and of less than about 25% with the NaI(Tl) detector.

Positive Autoradiographic Results

The autoradiograph of the leading edge of the blade section used to make the graphs in Fig. 2 is shown in Fig. 1. Four definite cracks are visible in Fig. 1 and are indicated by black exposure lines perpendicular to the leading edge. The kryptonated turbine blade was painted white to make the darkened portion of the film visible and then covered with liquid film emulsion.

DEVELOPMENT OF KET TECHNIQUE

The work described in the preceding paragraphs completed the requirements of the USAF contract. However, Industrial Nucleonics personnel believed that positive and fast results might be obtained through the use of a process that we have chosen to designate as the KET. Therefore a project was initiated under support from Industrial Nucleonics funds.

Since the ability of kryptonated materials to retain their activity with time is an undesirable property for the present purpose, the time and temperature at which turbine-blade samples were exposed to a ^{85}Kr atmosphere were varied to induce only diffusion into microcracks and adhesion to the surface. Air is known to be adsorbed onto metal surfaces exposed to the atmosphere and to outgas from them if the metals are placed in a vacuum. Since O_2 and N_2 are slowly released when the concentrations of these gases are decreased outside a metal, as in a vacuum chamber, the possible subsequent take-up of ^{85}Kr and its release on exposure to normal atmospheric gases were of interest.

The KET is a process for examining defects (cracks, pores, etc.) in materials by use of radioactive krypton gas. It involves degassing the material, placing it in a radioactive krypton atmosphere at or below room temperature, removing the material from the krypton atmosphere, and rapidly measuring the surface radioactivity before the krypton has escaped from the defects. In this case, krypton is strongly adsorbed but is hardly absorbed at all. Thus the KET treatment is quite different from kryptonation, in which absorption is the dominant process. Absorption, of course, requires temperatures appreciably above room temperature.

Radioactive Emission Results

The effect of using the technically simple KET conditions was determined in an experimental examination of six turbine-blade sections. The conditions of the experiment included were a pressure of 67 cm Hg, a temperature of 22.2°C, and an exposure of 1 hr. Count-rate measurements vs. time (Fig. 3) indicate an almost classical diffusion behavior with time in contrast to that of Kryptonates. Kryptonates retain their activity with time, but these samples had lost 90% of their initial measured activity after 9 hr. While the loss rate was too great to permit time for autoradiography to determine if the cracks still concentrate the activity under the new experimental conditions, further development of an electronic device with fast response time would allow such a determination. The loss rate

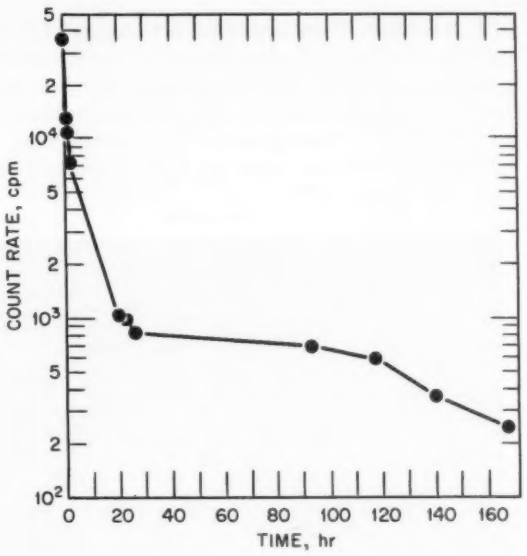

Fig. 3 Outgassing measurements. The KET parameters: time = 1 hr, temperature = 22.2°C, pressure = 66 cm Hg.

does indicate that the residual activity in a blade after 24 hr would be reduced by a factor of almost 50. Thus, if crack detection could be done immediately after KET treatment, very little residual activity would remain in a turbine blade by the time it was ready for installation in an engine. However, count-rate measurements on these blades from the time the KET vessel was opened indicate that the initial activity levels for the first 3 hr were greater than those constant levels which result from kryptonation. The kryptonation process produced blades with a constant count rate of 7000 cpm, which is comparable to the count rates 3 to 4 hr after the KET vessel was opened (Fig. 3).

Electronic Results

The cracks indicated in the autoradiograph were not detected by the small nuclear detectors because the amount of activity in the defects was not sufficiently greater than the external activity. Hence an electronic detector with significantly better spatial resolution was deemed necessary. The area of the crack was less than 1% of the total area examined. A detection system with a resolution capability of one detection element per area of interest was desirable, but the high spatial resolution required for this detection was not available at Industrial Nucleonics. Therefore several potentially useful detection devices were tested which would offer

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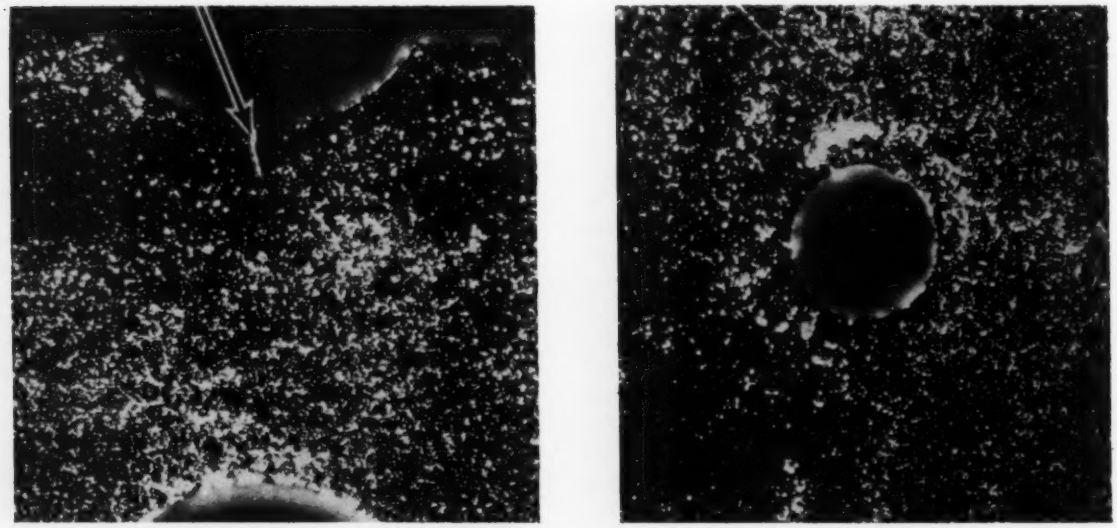
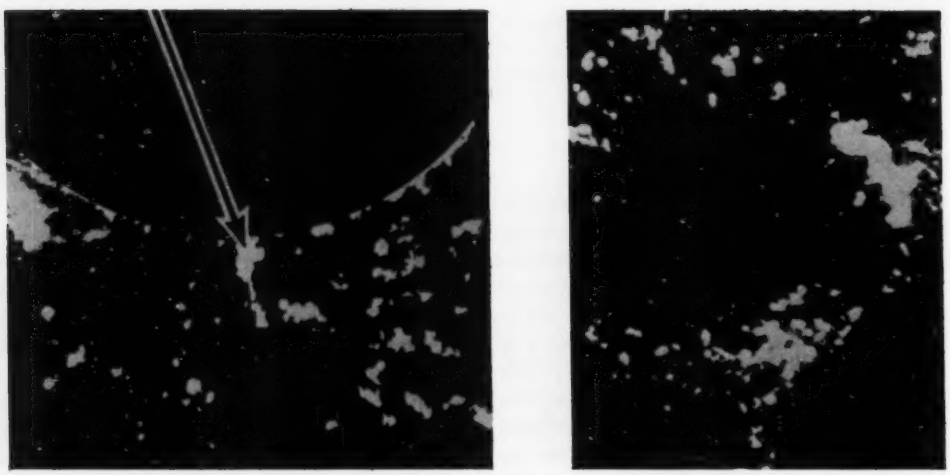
(1) the speed of an electronic detector and (2) the spatial resolution of autoradiography. Several cracked specimens that had been kryptonated were examined with a low-level light intensification system recently developed by Westinghouse. The system used a phosphor conversion layer for high efficiency ^{85}Kr beta detection, coupled with a fiber optics bundle for high spatial resolution. The individual fibers have a 16-μm diameter.

The time requirement and spatial detail of autoradiography and electronic imaging are compared in Figs. 4 and 5. The sample used in Fig. 4 was a



(a) (b)

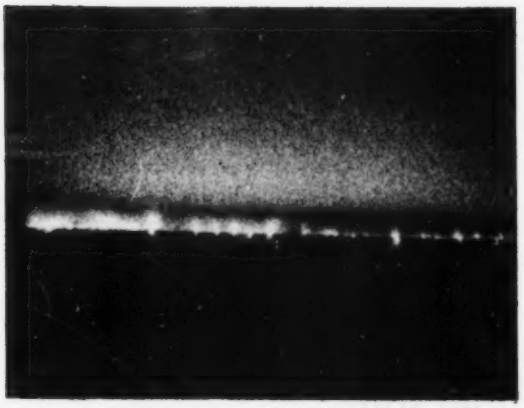
AUTORADIOGRAPHS FROM 3-hr EXPOSURE



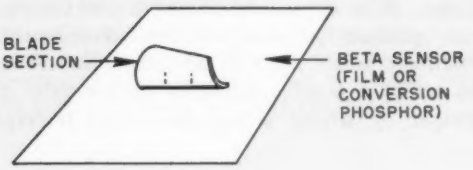
(c) (d)

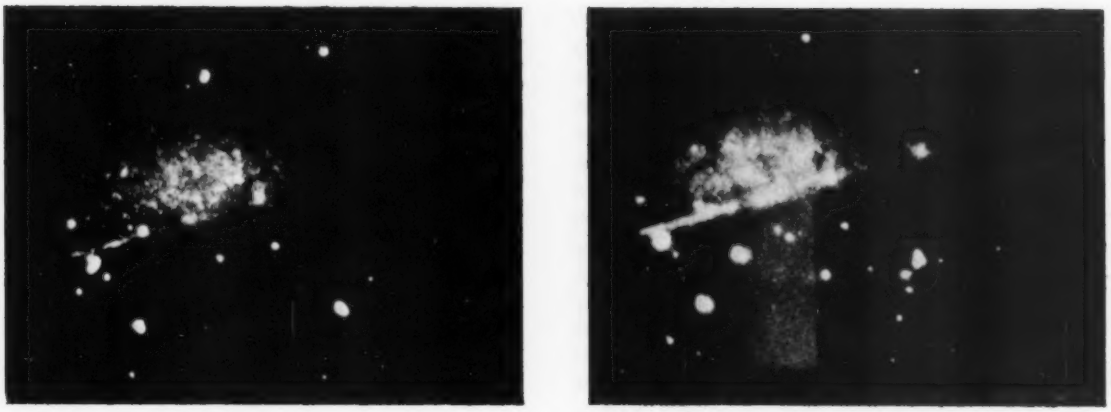
ELECTRONIC IMAGING RESULTS FROM 20-sec EXPOSURE

Fig. 4 Comparison of crack detection by autoradiography and by electronic imaging; (*b*) and (*d*) compare details of hole periphery.



AUTORADIOGRAPH FROM 3-day EXPOSURE





(a) (b)

ELECTRONIC IMAGING RESULTS FROM (a) 10- AND (b) 20-sec EXPOSURES

Fig. 5 Comparison of leading-edge crack detection by autoradiography and by electronic imaging.

kryptonated aluminum plate that had been cracked but not broken by stress fatigue as part of another program. The plate has a flat surface with two holes drilled through it; (*b*) and (*d*) in Fig. 4 compare the detail of the hole periphery. The autoradiograph in Fig. 4 required 3 hr for exposure; the electronic process required only 20 sec for the results shown. Examination of the cracked region and its periphery indicate that a close correspondence between the two detection methods exists. Figure 5 gives the results of an end-on measurement of a kryptonated turbine-blade section by electronic means and a comparable autoradiograph. (The electronic device allows comparison of its output, which is in pictorial form, with autoradiographic results. The electronic device is also about 10^5 times as fast as autoradiography.) The turbine-blade section used had a constant count rate of about 7000 cpm, which is comparable to that found 3 hr after a KET treatment (Fig. 3). The diagram in Fig. 5 shows the sample positioning used in both sensing methods. Owing to the convex shape of the turbine blade's leading edge, no pertinent information was obtained perpendicular to the leading-edge axis by autoradiographic or electronic means. The intense "point" regions in both the autoradiographs and electronic imaging methods indicate cracks. A white "halo" above the leading edge in all photos of Fig. 5 is due to ^{85}Kr activity located on the sides of the blade.

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Industrial Nucleonics has obtained photos showing crack extension by coating the entire leading-edge surface with liquid film emulsion. The exposure time for autoradiography at these activity levels is about 2 days. Since the sample placement used for Fig. 5 is not optimum for subsequent crack discernment, the electronic sensor could be advantageously redesigned to realize a possible geometric improvement in detection by sensing around the curved leading-edge surface. A beta detection system can be provided for such an inspection system. (FJM)

SUGGESTED REFERENCE

J. E. Carden, Radio-Release in Review with Special Emphasis on ^{85}Kr Clathrates and Kryptonates, USAEC Report ORNL-IIC-18, July 1969.

Neutron Radiography*

Six years ago, only two or three laboratories were working in the field of neutron radiography,[1] but by 1969 there were more than 50 centers with neutron radiography facilities (Fig. 1). Most of the present application work in neutron radiography uses a nuclear reactor as the neutron source.[2,3] However, there has been some development work on small-source neutron radiography,[4] and facilities using nonreactor sources are being set up in several centers, e.g., Argonne National Laboratory's facility at Idaho Falls (^{124}Sb–Be, 6 thousand curies), Euratom's facility at Ispra, Italy (neutron generator, 10^{11} n/sec), and the Marshall Space Flight Center at Huntsville, Ala. (Van de Graaff accelerator, 10^{12} n/sec). If neutron sources suitable for neutron radiography become available with a convenience and cost comparable to those of existing X- and gamma-ray sources, interest in the technique will spread rapidly. A parallel may be drawn between the development of neutron radiography and that of X radiography during the period 1900 to 1930, when adoption of the X-ray technique was slow until reliable and inexpensive X-ray sources became available. It should be emphasized, however, that the parallelism does not apply to the applications per se—the two techniques are complementary rather than competitive since they supply different types of information about the interior of an object being examined.

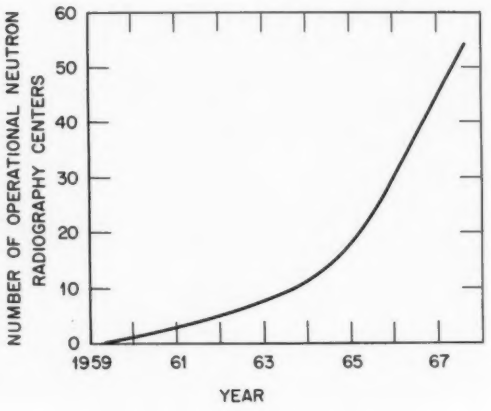

Fig. 1 Growth of neutron radiography.

Currently, most applications of neutron radiography are in the modern technical industries, such as nuclear energy, aeronautics, weapons technology, and space technology, but interest is developing in other fields, e.g., medical research.[5,6] With the eventual availability of small, inexpensive neutron sources, the variety of applications may become as wide as that for X radiography.

Three papers on neutron radiography are presented in this issue of *Isotopes and Radiation Technology* to show three phases of the work as it now exists. The first paper is on the possible application of ^{252}Cf, which fissions spontaneously to produce neutrons; the second is on the use of an accelerator; and the third is a description of a nuclear reactor facility. (MG)

*This introduction to the three neutron radiography papers that follow is taken from J. P. Barton, Foreseeable Application of ^{252}Cf to Neutron Radiography, in Californium-252, Symposium Proceedings, New York, N. Y., Oct. 22, 1968, James J. Barker (Ed.), USAEC Report CONF-681032, pp. 303-319, New York Metropolitan Section of the American Nuclear Society, 1969.

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Foreseeable Application of ^{252}Cf to Neutron Radiography*

By J. P. Barton†

Abstract: *The foreseeable future demand for ^{252}Cf as a neutron source for neutron radiography is analyzed in the light of the present rapid growth of application work in this field.*

It is predicted that if a decision to produce ^{252}Cf were taken, this neutron source material could be available about 10 years from now at the attractive cost of $250 for a 10^9-n/sec source (half-life 2.65 years). An assessment of the size of the anticipated demand for ^{252}Cf many years in the future requires, among other things, a look at the possibilities for small-source neutron radiography.

Foreseeable neutron-radiography techniques using small sources fall into three distinct categories: (1) thermal-neutron sources with direct-exposure radiography, (2) thermal-neutron sources with gamma-insensitive exposure, and (3) fast-neutron sources. The first category is, at present, the most likely application for transplutonium isotopic neutron sources. Gamma-insensitive exposure would be necessary for examination of highly radioactive objects, but present detection methods, such as the activation transfer process, have low efficiency and therefore require very intense neutron sources. Fast-neutron radiography would make good use of small isotopic neutron sources since there would be no waste of neutrons in moderation or collimation, but the techniques of fast-neutron imaging are, as yet, relatively undeveloped, and consequently the demand for fast-neutron radiography application work is unproved.

REQUIRED SOURCE CHARACTERISTICS

Physical Size

Thermal-neutron radiography is achieved by surrounding the small isotopic source of neutrons by a block of moderator. A 30-cm radius of ordinary water around the source is a convenient and fairly efficient moderating system. A neutron collimator is then inserted to the region of maximum thermal-neutron flux (which occurs near the central source). The collimator extracts the thermal-neutron beam and directs it to the object exposure position.

With the principle of the divergent collimator,[1] it can be shown that the source–moderator combination is required to provide the highest thermal-flux peaking in one small region, rather than a more voluminous but slightly lower thermal-flux distribution. The very small physical size of a ^{252}Cf source could possibly lead to an advantage in obtaining a peak thermal flux in a moderator close to the source. This is because of the small moderator displacement and the small absorption of diffusing neutrons in the source itself. However, in present practice we should note that the ^{252}Cf source is likely to be placed inside a container of steel or similar material, and the outer dimension of this metal encapsulation may be 2 or 3 cm so that the effective

*Excerpts from a paper presented at the Californium-252 Symposium, Oct. 22, 1968, New York, N. Y., sponsored by the New York Metropolitan Section of the American Nuclear Society. This updated version reprinted by permission of the author and of the proceedings editor.

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source size is not dissimilar to that of typical small (α,n) sources such as Am–Cm–Be.

For fast-neutron radiography the size of the source would be very important, and the high specific activity of ^{252}Cf should be a distinct advantage (Fig. 1). The geometric unsharpness in fast-neutron radiography is set by D/L, where D is the effective diameter of the source and L is the distance between source and object. Since the available neutron intensity at the exposure position is a function of $1/L^2$, for a given picture quality (geometric unsharpness), a geometrically small spontaneous-fission source of ^{252}Cf would be superior to a source of another type that had the same strength but was geometrically larger, e.g., ^{239}Pu–Be or ^{241}Am–Be. The magnitude of this effect may be stressed by noting that ^{252}Cf has a specific activity of 2.3×10^{12} n/sec per gram of material. This may be compared with the specific activity of subplutonic radioactive sources, which is about 10^6 n/(sec)(g).

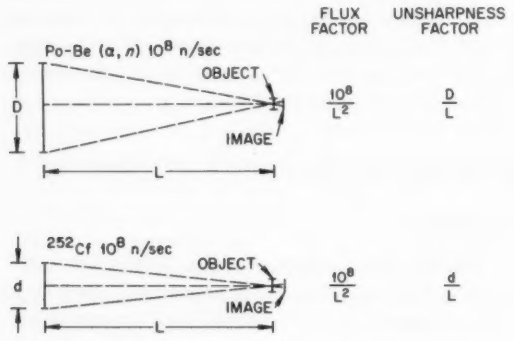

Fig. 1 Choice between (α,n) isotopic neutron source and spontaneously fissioning neutron source, each with an output of 10^8 n/sec, for fast-neutron radiography. The choice would be for ^{252}Cf because the unsharpness factor is smaller.

Another pertinent observation is that, if the neutron yield of a ^{252}Cf source is proportional to its size, then a large, intense source would have no advantage for fast-neutron radiography over a smaller, less intense source. This is because the smaller source would be placed nearer to the object than the large source for the same geometric unsharpness factor (Fig. 2). Thus one would choose the smaller source because of cost and convenience.

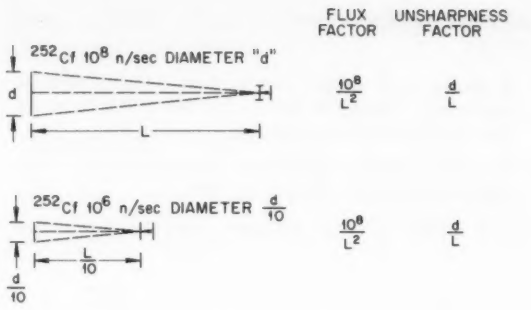

Fig. 2 Choice between ^{252}Cf sources of different sizes for fast-neutron radiography. The choice would be for the source of diameter $d/10$ because neutron radiographic capabilities would be the same for the two, but expense and handling convenience would be advantageous for the smaller source.

Neutron-Energy Spectrum

For thermal-neutron radiography the neutron-energy spectrum of the source is relevant in determining the ratio of peak thermal flux to source fast-neutron yield. It may also have an effect on the ratio of useful collimated thermal-neutron flux to unwanted fast-neutron interference at the exposure position, but, because the chosen detectors are predominantly sensitive to the thermal neutrons, this effect is of only minor importance.

Most (α,n) radioactive neutron sources yield neutrons in the range 1 to 5 Mev. The (γ,n) source ^{124}Sb–Be yields neutrons of about 25 kev energy, and there is evidence that this lower energy leads to a significant advantage for (gamma-insensitive) thermal-neutron radiography, with a gain, compared to (α,n) sources, of a factor of 10 in peak thermal flux per unit fast-neutron yield.

Californium-252 produces neutrons by spontaneous fission, and the mean energy of its fission spectrum is 2.3 Mev. The exact importance of this neutron-energy spectrum, slightly lower than the (α,n) spectrum, is unclear at this stage. However, it is very important for thermal-neutron radiography to note that peak thermal-neutron fluxes obtained with ^{252}Cf are a factor of 3 to 5 better than typical fluxes obtained with (α,n) sources of the same total fast-neutron yield.[2] This result could be due to a combination of factors, including the neutron-energy spectrum, source self-absorption, and moderator displacement.

Gamma Radiation

The contrast provided by neutrons in most applications of thermal-neutron radiography will be differ-

ent (or opposite) to that provided by X rays. Hence, for the direct-exposure method, gamma emission from the neutron source will tend to interfere and fog the neutron picture, and a low gamma/neutron output ratio is desirable. To date we have demonstrated that ^{241}Am–Be and ^{241}Am–^{242}Cm–Be have acceptable gamma/neutron ratios for practical direct-exposure neutron radiography and that ^{252}Cf will also prove acceptable.

The significance of gamma contamination in applications of fast-neutron radiography is uncertain at this time. Probably gamma interference will be less embarrassing because fast-neutron cross sections are of the same order of magnitude for different materials, whereas thermal cross sections change abruptly from one material to another. Fast-neutron and gamma-ray shadows will therefore not be in opposition. There is also the possibility that gamma-insensitive detection systems, such as the track-etch technique, may eventually be used in fast-neutron radiography.

REQUIRED SOURCE INTENSITIES

The source strengths of interest for applications of thermal-neutron radiography will be in the range 10^7 to 10^{12} n/sec (10 μg to 1 g). Small sources will require less shielding and will be more transportable, but their use will be limited to direct-exposure methods and will result in long exposure times or radiographs of low definition. For a particular application the source strength should be matched to the interrelated parameters (1) required detail visibility, (2) object thickness, (3) required collimation, (4) inherent unsharpness of the detection system, (5) detector efficiency, and (6) acceptable exposure times. Table 1, where the experimental results obtained with various neutron sources

Table 1 Radiography Characteristics of Small Neutron Sources

Source-strength yield, fast neutrons per 4π solid angle per sec	Sources evaluated	Thermal-neutron-flux collimator input, n/(cm^2)(sec)	Typical collimation ratio, input diameter x length	Beam intensity at collimator output, n/(cm^2)(sec)	Typical exposure
10^7	^{241}Am–Be	10^4	3 x 100	1	3 days, scintillator + fast film
			10 x 100	10	9 hr, scintillator + fast film
10^9	Small neutron generator (SAMES B.S. 1), ^{241}Am–^{242}Cm–Be	10^6	3 x 100	10^2	1 hr, scintillator + fast film
			10 x 100	10^3	6 min, scintillator + fast film
10^{11}	D-T neutron generator (SAMES)	10^8	3 x 100	10^4	0.5 min, scintillator + fast film
			10 x 100	10^5	2 min, scintillator + fine-grain film
	Reactor at 1 kw (Juggernaut, Argonne)	10^9	3 x 100*	10^5	2 min, scintillator + fine-grain film
	Reactor at 200 kw (Juggernaut, Argonne)	2×10^{11}	3 x 100*	2×10^7	1 min, direct-exposure gadolinum foil + fine-grain film (0.0005 in.) (Kodak Single R)
	High-quality reactor facility (Melusine 4 Mw or Siloe 15 Mw, Grenoble)	10^{13}	1 x 100	10^8	Sufficient intensity and collimation to penetrate through thick object and still use thin activation-transfer foils and very-fine-grain film

*The Juggernaut beam collimator is a simple parallel-sided hole penetrating the reactor shield. It is not cadmium lined. All other collimators quoted are lined with a material opaque to thermal neutrons (cadmium or boron) and are of the divergent type.

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are summarized, gives an insight into the relations among source intensity, radiographic quality, and exposure time.

Some experimental observations that should be taken into consideration are:

Range of Collimator Ratios: Small-source neutron radiography has been noted to work well over a certain range of collimator ratios, but, if very high degrees of collimation are attempted, the radiographic contrast depreciates significantly. This is probably due to the worsening ratio of thermal-neutron flux to interference radiation (fast-neutron and gamma radiation) as the collimation is increased. Collimator ratios in the range from 100 : 10 to 100 : 2 have given satisfactory small-source radiographs, and this range of collimation is satisfactory for many applications.

Collimator Optimum Aperture: We should also note that, having chosen the collimator ratio, the actual dimensions of the collimator are to be carefully selected. An inlet aperture which is too small might mean that the corresponding length would be too small—leading to interference problems from source radiations and to divergence-angle problems for radiography of sizable objects. An inlet aperture which is too large would mean that the collimator is looking at the thermal-neutron flux over a region significantly larger than the region of thermal-flux peaking, and it could mean a depression of the source thermal flux due to moderator displacement and neutron absorption. An optimum inlet-aperture dimension appears to be about 7 cm.

Geometric Unsharpness: The geometric unsharpness, U, in microns, which is, of course, a function of the collimator of length L and inlet aperture A, is rather better than that predicted from the simple geometrical relation

$$U = 10^3 \frac{(S + t/2)A}{(L + t/2)}$$

where t is the thickness of the object and S is the separation between object face and imaging screen. For example, with a collimator 1000 mm long and 70 mm inlet aperture and an object 8 mm thick separated from the imaging screen by a distance of 1 mm, we calculate the geometric unsharpness to be

$$U = 10^3 \frac{(1 + 4)\,70}{(1000 + 4)} \simeq 350\,\mu$$

and find that high-contrast details in the object can be distinguished when separated by this or even somewhat smaller distances.

Detector Efficiencies: A very efficient neutron imaging system consists of a ^{6}LiF–ZnS scintillator adjacent to a fast light-sensitive film. The scintillator gives rather a grainy image but requires a neutron exposure of only about 5×10^5 n/cm^2 with Kodak Royal Blue film. Slower film, such as Ilford HP3, gives an image with better inherent resolution for a neutron exposure of about 5×10^6 n/cm^2, while gadolinium-foil converters used in conjunction with medium-speed X-ray films give very-fine-grain neutron radiographs with exposures upward from 5×10^7 n/cm^2.

Combined Relations for ^{252}Cf: These figures may be used to calculate the required source intensity–exposure time relation. Experiments show that, for a typical thermal-neutron radiography arrangement using a ^{252}Cf neutron source (light-water moderator, divergent cadmium-lined collimator) the required exposure time, T, in minutes, is related to the ^{252}Cf source strength, S, in neutrons per second, by

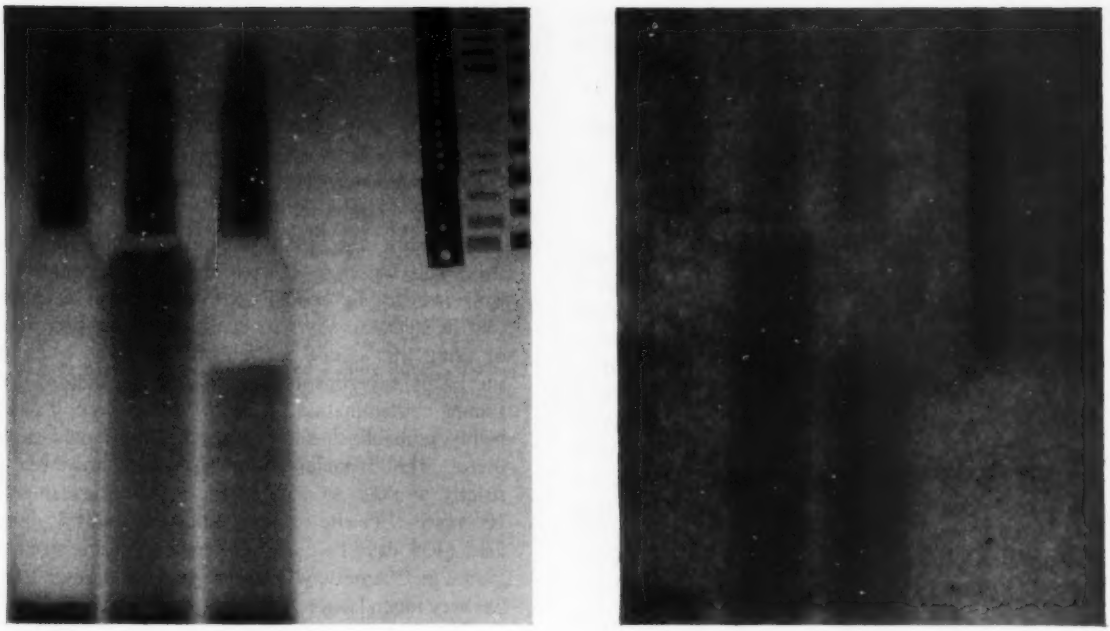
$$T = 40 \left(\frac{L}{A}\right)^{-2} \frac{D}{S}$$

where D is the neutron dose of the chosen imaging method (e.g., 5×10^6 n/cm^2) and L/A is the collimator ratio (L the length and A the inlet aperture). For example, using a medium-speed detection system with a D value of 10^7 n/cm^2 and a collimator system with a medium ratio, i.e., 100 : 7, a source strength, S, of 10^9 n/sec will provide neutron images with exposure times of

$$T = 40(100/7)^2 \times 10^7/10^9 = 80 \text{ min}$$

Examples of ^{252}Cf Radiographic Quality: Figure 3*a* shows a thermal-neutron radiograph of three bullets enclosed within lead walls of 6 mm total thickness. The radiograph clearly shows the extent of explosive loading within each bullet. In the top corner of the radiograph is an object-image-quality indicator.[3] The original radiograph negative shows plastic-wire visibility down to 0.015 in., and holes in the cadmium could be distinguished down to 0.010 in. separation for 0.010 in. diameter. The radiograph was taken with a ^{252}Cf source of 3.4×10^8 n/sec yield. The moderator was water; the collimator, cadmium, had a 7.6-cm aperture and a 10^6-cm length; and the imaging system was gadolinium foil (50 μ) with Kodak AA film. The exposure time for this radiograph was 64 hr.

Figure 3*b* shows the same object and the same arrangement, except that this time the imaging system was ^{6}LiF–ZnS scintillator used with Kodak Royal Blue



(a) (b)

Fig. 3 Neutron radiographs taken with a small ^{252}Cf neutron source (140 μg). They show the extent of explosive powder loading in three bullets enclosed in 6 mm of lead. Figure 3*a* was taken using gadolinium-foil neutron conversion, and Fig. 3*b* with LiF–ZnS scintillator conversion.

film and the exposure time was only 20 min. Clearly, a small ^{252}Cf source (such as the 140 μg of these experiments) could be useful for some applications, giving quite fine-grain radiographs of fairly thin objects in long exposure times, or rather coarser grain radiographs in short exposure times.

COMPETITION AMONG OTHER NEUTRON SOURCES

Neutron Generators

Neutron generators are now available and competitive with radioactive sources[4,5] over a wide range of neutron yields, e.g., 10^8 to 10^{12} n/sec. A typical low-yield neutron generator would be a low-voltage (150 kv) sealed-tube *D–T* device comparable in size, complexity, and cost with a small X-ray unit. The intermediate range typically would be a continuously pumped high-voltage set (150 to 600 kv) with replaceable tritiated targets. The useful life of these targets is at present about 20 hr for operation in the range from 5×10^{10} n/sec, and the replacement cost is about $100. At least one firm is offering a sealed-tube system with an expected life greater than 100 hr at 10^{11} n/sec output. Yields of 10^{12} n/sec and greater could at present be provided by Van de Graaff machines. Small cyclotrons are also available,[6] with the advantage that the tritium target problems may be avoided by using high voltages and alternative target reactions, such as D–Be. The cost of a small "tabletop" cyclotron is about $250 thousand.

Assuming that the costs for a given neutron yield and useful lifetime are comparable, it seems probable that some applications will call for neutron generators, while other applications will call for isotopic sources. The choice between neutron generator or isotopic source will probably depend on the importance of some of the following considerations:

1. Source operation for long periods (e.g., for lengthy routine work or long transfer radiographs) with 100% reliability.
2. Periodic source shutdown (e.g., to arrange sample).
3. Transportability and housing considerations.

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4. Use of source for other purposes (e.g., some neutron generators may be used as pulsed sources for activation analyses, neutron physics research, etc.).

5. Capital cost of the system.

6. Running costs (e.g., target changes, maintenance, depreciation).

Transplutonium Isotopic Sources

Americium-241-Be appears to be an attractive neutron source for neutron radiography, particularly when a direct-exposure portable source is required. The associated gamma radiation is soft and therefore easily filtered, thus allowing improved radiographic quality and easy gamma shielding for transportation. A 40-curie source (10^8 n/sec) would require about 500 lb of shielding during transport, and a smaller source would, of course, have lighter shielding but would provide the same modest quality of radiography if lengthy exposure times are acceptable. The half-life (460 years) causes no inconvenience, and the present cost ($3 thousand for a 10^7-n/sec source) is acceptable. Americium-241 has just started to become available in fairly large amounts and will probably become more abundant in the future. Unlike[7] some other transplutonium elements, ^{241}Am does not rely on a high-flux isotopic reactor for fabrication because it is generated spontaneously by burnup of plutonium in power reactors.

The neutron yield of a ^{241}Am-Be source may be increased by a factor of about 100 by irradiation of the source in a reactor neutron flux.[8] However, the resulting source ^{241}Am-^{242}Cm-Be has a half-life of only 163 days (compared with 460 years for ^{241}Am). This source may sometimes be preferred—for example, where activation-transfer neutron radiography is to be performed or where high-beam intensity is required for another reason. Recent experiments at Argonne National Laboratory using an Am-Cm-Be source (10^9 n/sec yield) have shown that direct neutron radiography is possible over a suitable range of collimation and that transfer radiography is possible for a 12- by 120-cm collimator used with dysprosium activation-transfer foil and the fast KK type X-ray film.

Another possible source for neutron radiography on which we hope to experiment is ^{244}Cm-Be. This material has a half-life of 18.1 years and a gamma/neutron ratio about 10% that of ^{252}Cf.

Subplutonium Isotopic Sources

The high gamma activity associated with the ^{124}Sb-Be neutron source not only makes direct-neutron photography impracticable, with consequent loss of efficiency, but also necessitates heavy and expensive shielding arrangements as part of the facility. Such systems would therefore not be considered convenient or portable. However, for inspection of radioactive objects, where hot-cell techniques or elaborate shielding systems may be necessary,[9,10] these objections do not arise.

Probably the most inexpensive and practical short-half-life small source for direct-exposure neutron radiography would be ^{210}Po-Be. Curiously, no successful experiments to demonstrate the possibility of using this source are known, but, in view of the successful experiments with ^{241}Am-Be and the low gamma contamination of the ^{210}Po-Be source, it seems highly probable that a polonium source would prove useful. The financial advantage would, however, be strictly limited to short-term applications since a 10^7-n/sec ^{210}Po-Be source would cost about $5 hundred (139 days half-life), whereas the same strength source in ^{241}Am-Be would cost only $3 thousand and last very much longer.

Other potentially competitive sources that may become available[11] include ^{227}Ac (21.8 years half-life) and ^{228}Th (1.9 years half-life). These sources can be produced by irradiation of radium in a nuclear reactor, followed by chemical separation. Very reasonable costs for sources in the range from 10^9 to 10^{10} n/sec have been envisaged.

CONCLUSIONS

There is a possibility of a demand 10 years from now for small radioactive neutron sources for neutron radiography. The main foreseeable advantages for ^{252}Cf would be:

1. The economic advantage of an inexpensive source with a convenient half-life.

2. The ability to obtain a high peak thermal flux for a given fast-neutron yield.

3. The possibility of working with an effective point source for fast-neutron radiography.

At a cost of 50 cents per microgram, a demand for ^{252}Cf could possibly be anticipated for

1. A large number of small sources (10^8 to 10^9 n/sec) for transportable thermal-neutron radiography systems.

2. A small number of large sources (10^{10} to 10^{11} n/sec) for activation-transfer neutron radiography in hot cells.

3. Sources for fast-neutron radiography applications that may include either industrial or medical work.

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Low-Voltage-Accelerator Neutron Radiography

By Frank A. Iddings*

Abstract: *A simplified system for producing neutron radiographs using a low-voltage-accelerator neutron source is presented. The system produces usable neutron radiographs in 1 min for samples such as electronics, ammunition and explosive devices, and other small specimens. Deuteron beam currents between 3 and 4 ma permit the necessary high neutron outputs, yet allow long target lives.*

Until isotopic neutron sources with high neutron output (such as californium promises to be) become available, only accelerators offer the combination of neutron output, low cost, and portability necessary for industrial utilization. Accelerators with a high-level deuteron beam (4 ma) at 150 kv are available with neutron yields greater than 10^{11} n/sec at a price less than $25 thousand. While nuclear reactors with fluxes 10^5 times those available from accelerators will produce the highest quality pictures, useful field work will be the domain of the small accelerators until sufficient quantities of ^{252}Cf are available.[1–3]

SYSTEM DESIGN

The system presented here is simple but adequate for industrial use, just as X-ray machines are used for X radiography although radioisotopes are available. Extension of this work to larger and more sophisticated samples should be possible. The objectives of

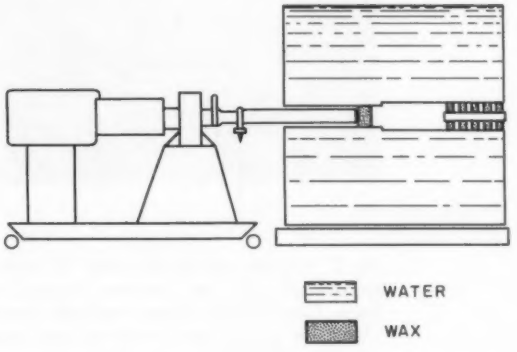

Fig. 1 Accelerator and shield arrangement for neutron radiography.

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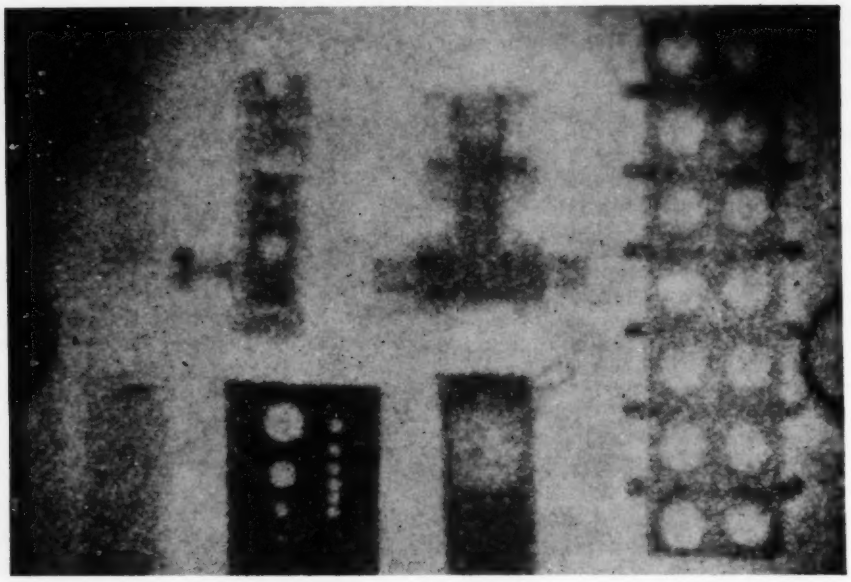
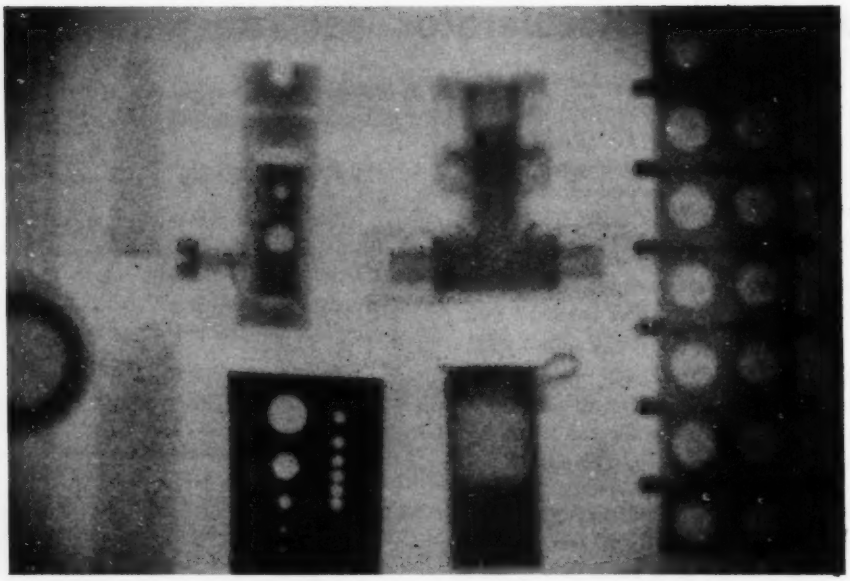


(a)



(b)

Fig. 2 Neutron radiographs using ^{3}H target, age 5 min at 3.5 ma; LiF/ZnS screen, source-film distance 60 in. (*a*) 6-sec exposure, Polaroid Type 57 film (ASA 3000). (*b*) 60-sec exposure at 4.2-ma beam, Polaroid Type 52 film (ASA 400). Specimens, left to right: rifle cartridge; stainless-steel capsule with nylon setscrew over a 0.020-in.-thick cadmium plate drilled with a series of $^{3}/_{16}$-, $^{1}/_{8}$-, $^{1}/_{16}$-, $^{1}/_{32}$-, and 0.024-in. holes as well as $^{1}/_{16}$-in. holes separated by different distances; coaxial T connector over plastic vial half-filled with water; and Bakelite terminal strip. Note that in *b* the $^{3}/_{1000}$-in. separation of $^{1}/_{16}$-in. holes in cadmium is clearly visible.

speed, low cost, simplicity, and good quality in neutron-radiograph production are satisfied by the design presented.[4]

The accelerator (Accelerators Inc. Accelerator I model) yields $>3.1 \times 10^{11}$ n/sec from a 5-curie ^3H target with a 4-ma deuteron beam and a 150-kv accelerating voltage. A drift-tube extension of 30 in. allows shielding and collimation of the isotropically produced 14-Mev neutrons. This shielding and collimation can be conveniently effected by use of a cubic water tank 6 ft on an edge (Fig. 1). A 6-in.-diameter pipe through the center of the tank forms the accelerator-drift-tube access and produces the 14-Mev neutron beam.

A 2.5-in.-thick plug of paraffin in front of the water-cooling jacket in the target produces a near-optimum thermal-neutron flux from the 14-Mev neutron beam and shield-scattered neutrons. Installation of smaller diameter tubes inside the 6-in.-diameter pipe yields better collimation and better picture quality. For the radiographs shown in Fig. 2, a 3-in.-diameter tube, supported by "donuts" of paraffin, was used. Additional paraffin moderator decreases the fast-neutron flux and at the same time decreases the thermal-neutron flux. The 14-Mev neutron component of the beam produces little loss in picture quality in the system used.

The LiF/ZnS (Nuclear Enterprises, San Carlos, Calif.) imaging screen combined with Polaroid Types 52 and 57 film gives the most convenient system for radiograph production.[5] Gadolinium screens coupled with radiographic film give slightly better pictures but with considerably less convenience and a large increase in exposure time. The LiF/ZnS screen mounted on a Polaroid Model 500 film holder proves to be a handy imaging system. The screen is mounted so that it can be pressed against the film or dropped away and still be light-tight.

Specimens are placed as close to the aluminum backing of the LiF/ZnS screen as possible. At distances of about 60 in. from the accelerator target, exposures are 6 to 10 sec or 45 to 90 sec for Polaroid Type 57 or 52 film, respectively. As the ^3H target is depleted and neutron output drops, exposures may take 10 times as long. The ^3H-target yield drops an order of magnitude at 4.0 ma beam current after about 4 hr use (Fig. 3).

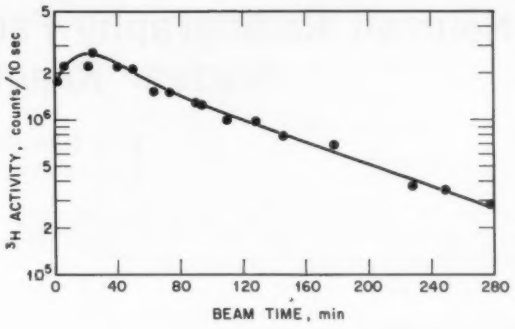


Fig. 3 Tritium-target depletion in a 4-ma deuteron beam.

NEUTRON RADIOGRAPHS

Figure 2 (*a* and *b*) illustrates the quality of results obtained with Polaroid Types 57 and 52 film, respectively. These radiographs also show the types of samples to which the system may be applied. Small electronic components, ammunition, and manufactured materials, in which a low-atomic-number part of interest is inside a high-atomic-number material, can be successfully radiographed. (MG)

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Neutron Radiography Facility at University of Texas Nuclear Reactor Laboratory*

By G. D. Bouchey and S. J. Gage†

Abstract: *The University of Texas neutron radiography facility, which is installed in a vertical beam tube from the 250-kw TRIGA reactor, is described.*

APPARATUS

Neutron Source

The neutron beam used for neutron radiography at the University of Texas is extracted from the TRIGA Mark I nuclear reactor located on the Austin campus. This reactor is capable of routine operation at a steady-state power level of 250 kw (Fig. 1), at which

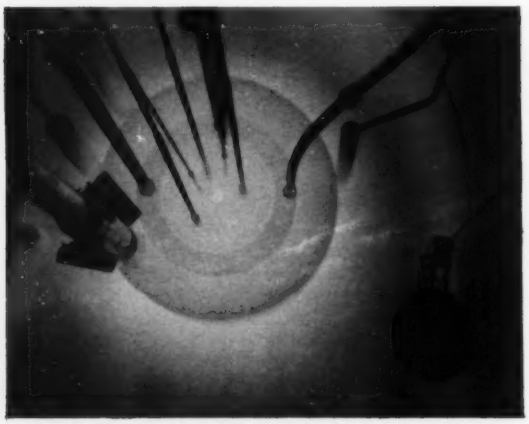

Fig. 1 The University of Texas at Austin TRIGA Mark I nuclear reactor during 250-kw steady-state operation, with beam tube resting on top surface of the reflector.

level the average thermal-neutron flux in the core is approximately 5×10^{12} n/(cm^2)(sec). In addition to steady-state operation, the reactor can generate power pulses of approximately 20 msec duration, with a peak power of 250 thousand kw and a total integrated power of ~15 thousand kw-sec. The reactor core consists of 62 cylindrical clad U-ZrH_2 fuel elements surrounded by a 12-in.-thick annular graphite reflector.

Beam Tube

The reactor core is located in a tank filled with ordinary water, 21 ft below ground level; thus a conventional horizontal beam port is not available for radiography. Instead, an air-filled assembly was constructed from aluminum pipe to provide a vertical beam tube (Fig. 2). The beam tube normally rests on the reflector that surrounds the reactor core. The lower portion of the tube consists of a 6-in.-ID by 6-ft-long section with a 2-in.-diameter lead plug attached to the bottom end. The lead plug decreases the gamma radiation in the beam to an acceptable level. The upper

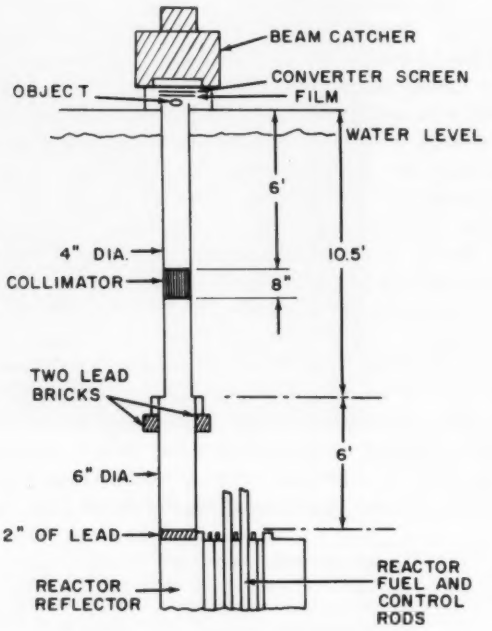

Fig. 2 Vertical-beam-tube assembly.

*Extracts from paper presented at Seventh Symposium on Nondestructive Evaluation of Components and Materials in Aerospace, Weapons Systems and Nuclear Applications, San Antonio, Texas, April 23–25, 1969.

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section of the beam is a 4-in.-ID by 10.5-ft-long tube, which extends above the water surface. All joints are welded, and the entire assembly is watertight. Because of the buoyancy of the tube, two 25-lb lead weights are attached to it to prevent its floating.

Collimator

The quality of the radiographs obtained depends strongly on the degree of collimation of the beam. The collimator for this facility was chosen to give an acceptable compromise between good-resolution radiographs and collimator cost; it consists of a bundle of 304 stainless-steel tubes inserted 6 ft from the top of the tube (Fig. 2). The tubes are 8 in. long and have an outer diameter of $\frac{1}{8}$ in. and a wall thickness of 0.012 in. The total cost of the uncut tubing was approximately $180. Figure 3, a radiograph of a 20-mil-thick cadmium sheet with 20-mil-diameter holes spaced at intervals of 30.4, 19.0, 9.0, 4.8, 3.6, 2.0, and 1.2 mils, demonstrates the quality of resolution obtained.

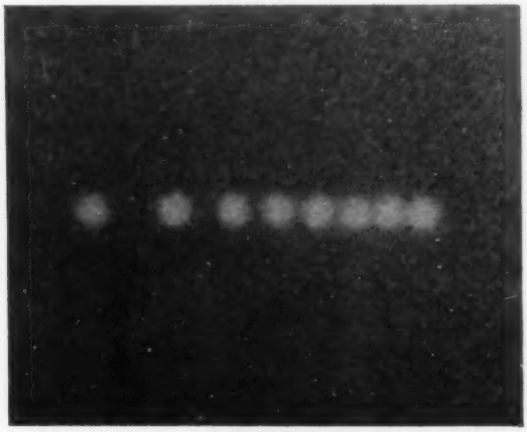

Fig. 3 Cadmium resolution test.

Radiography Assembly

A holder for the test object, the film, and the converter screen is located approximately 12 in. above the water surface (Fig. 4). The holder was designed so that objects and films may be changed without having to lower the reactor power level. A "beam catcher," constructed of neutron-absorbing materials, is located directly above the holder to eliminate any radiation hazard associated with the beam.

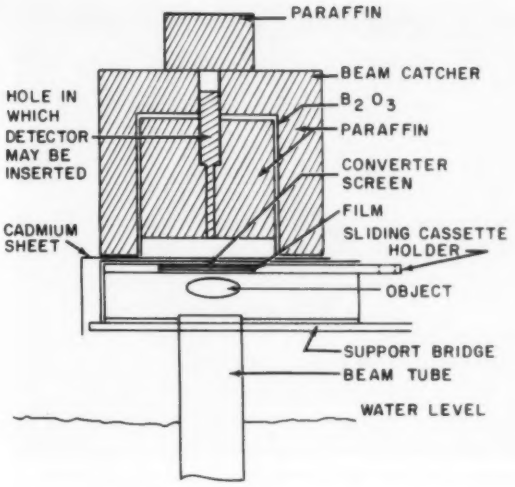

Fig. 4 Removable radiography terminal assembly and beam catcher.

PROCEDURES

During Steady-State Reactor Operation

The Texas facility at Austin provides an excellent and relatively versatile beam of neutrons for radiography. Radiographs of most objects, depending on the type of film and the thickness of the object, may be obtained with reasonably short exposure periods. For most work, Kodak Type AA X-ray film has proved satisfactory; for most objects it requires exposures of 1 to 5 min. When a finer grained film is desired, Kodak Type R film is used, but it is much slower and may require exposures of 30 to 50 min.

The quality of a radiograph can often be improved by varying the energy of the neutrons in the beam. Normally, with the beam tube resting on the graphite reflector, most of the exposure results from relatively slow (thermal, less than 0.5 ev) neutrons. A thin (20-mil) sheet of cadmium inserted in the beam removes most of the thermal neutrons but allows the higher energy neutrons to pass through. This, of course, increases the time required, but exposures are still quite reasonable—approximately 30 min for Type AA film. The neutron energy can be further increased by moving the base of the beam tube nearer the center of the reactor core. Radiographs using higher energy neutrons can give increased contrast for certain objects, particularly large thicknesses of materials with large scattering cross sections, such as water or steel. Figure 5*a* shows a radiograph of a relatively

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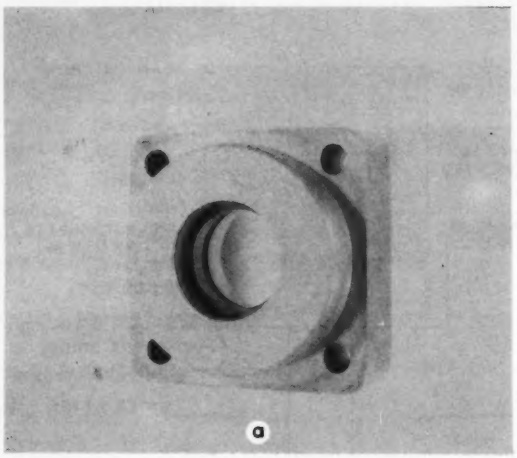

thick steel flange. The increased contrast in a radiograph which is obtained with the more energetic epithermal neutrons over that obtained with the usual thermal-neutron spectrum present during a steady-state operation is indicated by Fig. 5*b*. This increased contrast allows the inspection of a rubber bearing seal located inside the rather massive piece of steel.

During Reactor Pulsing

Neutron radiographs can also be obtained using a pulse from the reactor. The pulsing method is especially attractive for applications that require "stop action" radiography. Kodak Type AA film is suitable for a pulse of approximately 15 thousand kw-sec and 20 msec duration. Pulses of this type can be repeated about once every 10 min. (MG)

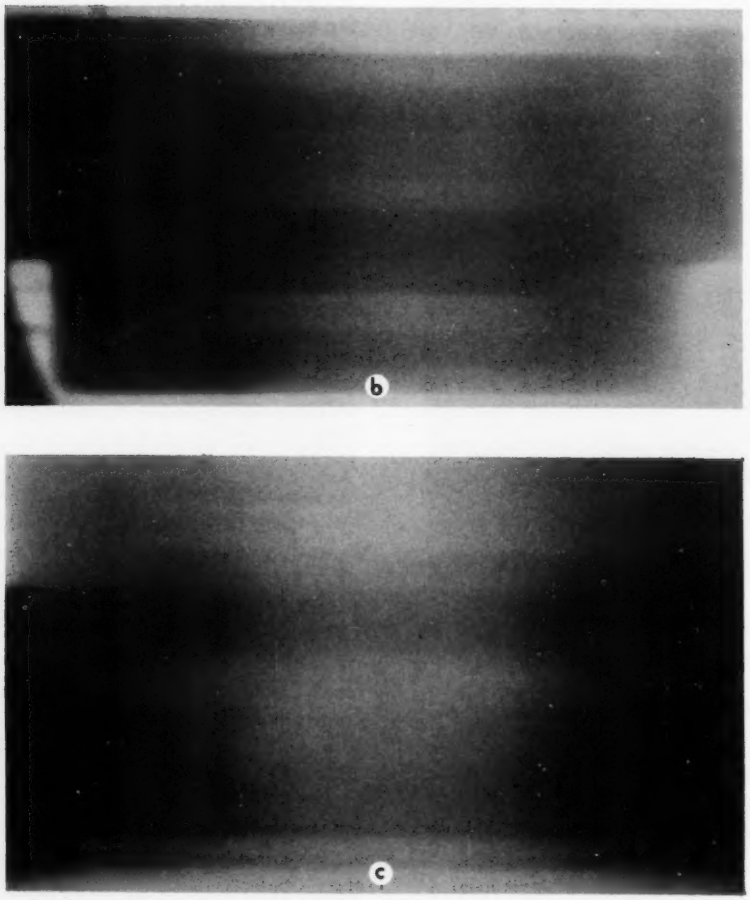

Fig. 5 (*a*) **Steel flange, and radiographs of it using** (*b*) **thermal neutrons and** (*c*) **epithermal neutrons.**

Neutron Meter Use in Soil-Moisture Measurements

A Report of a Survey Conducted by the U. S. Work Group on Nuclear Techniques in Unsaturated and Saturated Zones

By Wilford R. Gardner*

Abstract: *The answers to a questionnaire sent to 350 persons from which 200 responded have been correlated to provide up-to-date information on the use of neutron meters in soil-moisture measurement. Commercial instruments are used chiefly, and the number in use increases steadily. Field calibration remains a difficult problem. The method is in use for every hydrological problem that requires a soil-moisture measurement. A real need exists for research and development to increase the reliability of the meters in field use. Two bibliographic lists are appended to illustrate the scope of present work.*

At the first meeting of the International Work Group on Nuclear Techniques in Saturated and Unsaturated Zones held at the International Atomic Energy Agency (IAEA) in Vienna in March 1966, member states of the committee participating in the International Hydrological Decade (IHD) were asked to provide information concerning the use of nuclear techniques in hydrological investigations. Accordingly the corresponding U. S. Work Group (a subcommittee of the U. S. National Committee for the IHD) sent out a questionnaire concerning the use of neutron moisture meters to several hundred scientists and engineers believed to have interest in neutron moisture measurements. The questionnaire was patterned after one suggested by IAEA but was much more comprehensive. A copy is included in Appendix A.

Because of answers obtained from the previous questionnaires by the American Society for Testing and Materials (ASTM) (1962) and by the Hydraulic Division of the American Society of Civil Engineers (ASCE) (1964), more is known, perhaps, about the use of neutron meters in the United States than in many countries. The present questionnaire was circulated in order to bring the information up to date and to provide fullest possible cooperation with the IHD.

The mailing list was assembled from names provided by concerned federal agencies, the University Council on Water Resources (UCOWR), and several interested individuals. Undoubtedly many users and other interested persons were overlooked. Questionnaires were sent to approximately 350 people (with duplicates for colleagues who might otherwise be missed), and about 200 were returned. The individuals who responded appear to be representative of all the users in the United States. Most of the answers were received by Aug. 1, 1967 (some responses were still being received much later), and this report summarizes the status of neutron meter use to that date.

About one-fourth of the respondents had not yet used the meter but expressed an interest in information concerning its use. Because in some instances several respondents were probably referring to the same piece of equipment and in other instances some groups had several sets of equipment, the exact number of instruments in use could not be established. However, this uncertainty is probably not serious. This report does not purport to describe all aspects of neutron meter use, but rather summarizes the responses to the specific questions asked.

SUMMARY OF RESPONSES

Description of Neutron Meter Used

Only two or three of the instruments in use were custom built; the majority were standard commercial models. Most of these were supplied by Nuclear-Chicago† or Troxler Electronic Laboratories,‡ with instruments from these two companies represented almost equally. Although some information was requested concerning surface probes as well as depth probes, the results revealed that the surface probes were used almost exclusively in highway-engineering applications and not in hydrological problems. Therefore the information reported below is only for the depth probes.

Approximately 146 probes used ^{226}Ra–Be sources and 58 used ^{241}Am–Be. There were 3 ^{238}Pu–Be sources and 5 unidentified. Almost all of the ^{226}Ra–Be sources were in the 3- to 5-mc range, except for 10 which were 2 mc and a few which were 8 to 12

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†Nuclear-Chicago Corp., a subsidiary of G. D. Searle & Co., 333 E. Howard Ave., Des Plaines, Ill. 60018.

‡Troxler Electronic Laboratories, Inc., P. O. Box 5997, Raleigh, N. C. 27607.

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mc. Of the ^{241}Am–Be sources, 39 were 100 mc, 8 were 80 mc, 4 were 50 mc, and the remainder had other source strengths ranging from a low of 5 mc to a high of 200 mc. The three ^{238}Pu–Be sources had strengths of 5, 1, and 0.125 curies.

Some estimate of the rate of increase of the use of the neutron meters can be gained from Table 1, which shows the number of instruments purchased annually. The data in this case may be subject to the accuracy of the respondents' memories.

Although the trend fluctuates slightly from year to year, the annual acquisition rate is increasing linearly. Implications of this will be discussed later.

About 40% of the probes had the source at the end of the counter, with the remaining 60% placed at the center. The latter geometry seems to be increasingly preferred.

No instruments of foreign manufacture were reported. A recurrent rumor that one of the two major suppliers was going to discontinue the manufacture of its neutron meters was denied by a representative of that company.

Table 1 Neutron-Meter Acquisition by Years

Year	Number	Cumulative number
1956	2	2
1957	6	8
1958	18	26
1959	14	40
1960	14	54
1961	9	63
1962	20	83
1963	13	96
1964	23	119
1965	20	139
1966	25	164
1967 (part year)	20	184

Calibration

Laboratory Calibration. About 50 of the respondents made no laboratory or field calibration but used the manufacturer's calibration directly. Many of those who did carry out laboratory calibrations were concerned with instrument reliability and reproducibility rather than with obtaining a calibration curve for field use. The containers and materials used to construct calibration standards varied widely. The smallest was a box about 38 cm square by 76 cm deep. The largest was a box 150 cm on a side. Many respondents used 55-gal drums. Water was usually used as the hydrogenous material in combination with such "inert" materials as sand, soil, clay, or boric acid. Polyethylene, quartz sand and alum, and $CdCl_2$ were also used.

Accuracy figures reported varied from 0.5 to 3 vol.% at low water (hydrogen) contents and 0.1 to 2% at high water contents. There seemed to be no obvious correlation between accuracy and instrument geometry, source strength, or other instrument specifications, although this was not examined in detail.

Field Calibration. Most experienced users (as judged by years of ownership) relied upon calibration in the field. Some 55 respondents indicated this was their practice. Reference standards were containers of water, paraffin, or cadmium, in most cases. Frequent shield counts were also used. The necessity of field calibration seems to be borne out by the experience of those who have used the method as well as by the theoretical studies of Holmes[1] and Ølgaard and Haahr.[2]

Major problems encountered in the field calibration were mainly related to the problem of sampling the soil gravimetrically due to heterogeneities and stones. High soil salt, iron, and organic-matter content also presented problems. A nonuniform water-content distribution, whether due to layering or water uptake by plant roots, was also cited as a problem. Bound water in lateritic soils proved to be a serious drawback with one of the instruments because of a leveling out of the calibration curve and loss of sensitivity. Some instruments proved to be somewhat more temperature sensitive than desirable in the opinion of some respondents, although no specifics were given.

All field calibrations were carried out with gravimetric samples as the standard. The number of samples taken varied from 4 or 5 to as many as 500 in one case. Many workers found it convenient to take samples from the hole as the access tube is inserted into the soil. In some cases they feel that this is sufficient in their soils, although these samples are usually supplemented with samples from two or three other nearby holes. The accuracy quoted for the field measurements ranged from about 0.5 to 2 vol.%, with many workers feeling that it was usually about 1%.

Access Tubes

By far the most workers used aluminum access tubes, with steel being the second most popular material. One worker tried a polyvinyl chloride tube, which was found to reduce the count rate noticeably. Some of the steel tubes rusted out within 3 years in

saline soil, but many workers have had tubes installed in soil as long as 5 years with no serious problems.

The installation methods reported were as varied as the soils in which the tubes were installed. The most frequently reported procedure was to auger the hole, either by machine or by hand, prior to driving in the tube. The next most frequent procedure was to auger out the soil from within the center of the tube as it was being driven into the soil. In some cases where aluminum tubing was used, a steel tube was first driven in, removed, and then replaced by the aluminum tube.

Where other means failed, a larger hole was sometimes bored with a power auger and backfilled after insertion of the tube. Cable drilling machines and jetting rigs were also reported as having been used. The most serious problems encountered were in drilling holes in stony soils and in inserting tubes to depths beyond about 25 ft in any soil. In many cases the bottom end of the tube was plugged after insertion to prevent entrance of water. Occasionally a desiccant was put in the bottom of the tube.

Field Application

The method was reported as being used for almost every hydrological application requiring a soil water-content measurement. This included evapotranspiration, irrigation, infiltration, deep percolation, drainage, and plant-water uptake. A list of the publications reported by the respondents is included in Appendix B and gives a representative sample of the types of applications and the way in which they were carried out.

Extent of Use

Information submitted represented up to 10 years' experience with this method on the part of some respondents. The average for all respondents reporting actual experience was 5 years. For whatever the terms mean, 55 respondents use the method "continuously," 56 use it "frequently," and 23 use it "occasionally." No respondent indicated having abandoned it for some other method, although some have not adopted it wholeheartedly.

Overall Evaluation of the Method

Of the responses to this question, 105 indicated the accuracy to be satisfactory and 7, unsatisfactory. One hundred ten were satisfied with the reliability and 6, unsatisfied. Ease of maintenance was not a problem for 90 but was unsatisfactory for 20.

Repair problems evoked considerable response. Thirty respondents found this to be a serious problem, and some suggested that one should own enough instruments so that at least one was operable at any given time if the data were important. Seventy respondents indicated that repairs were a minor problem, 19 found them no problem, and 2 reported a hopeful "not yet." There seemed to be an inverse correlation between the proximity of the repair service and the degree to which repairs were considered a problem. Many locations reporting no problem also revealed that they had their own repair facilities. Some respondents found the service good, but costly, and others experienced considerable downtime because of their distance from the repair service. One contented user found a local TV repairman who had been able to cope with most maintenance problems to date.

Developmental Needs

Many users had suggestions for improvements they would like to see made. These can be roughly divided into two categories. The first is in the design of the instrument to allow measurements that are not now possible or cannot be made with adequate precision. Such developments would include devising ways to make better surface measurements and to give better depth resolution. Both larger and smaller sampling volumes were requested. These problems are inherent in the nature of the neutron-scattering process and will require the attention of physicists who understand the problems.

The second type of improvement has to do with the engineering of the instrument and associated equipment for more convenient or efficient field use. In this category are requests for easier calibration procedures and easier means of lowering and raising the probe to predetermined levels. An almost universal complaint was that the present instruments are not rugged enough, particularly at vulnerable points such as the cable connections. Some users would like to see an automatic lowering device and a strip chart or other recorder that would also integrate the water content as the probe was moved. Lighter weight would be welcomed by all users.

Typical general comments ranged from ". . . it is much superior to gravimetric sampling. . ."; ". . . as good as any method, best for our purpose"; ". . . satisfactory for field research but not precise enough for research"; to "not good enough, but best there is. . ." and ". . . we use gravimetric sampling if we really want to know. . .".

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CONCLUSIONS AND RECOMMENDATIONS

It seems apparent that, after more than a decade of experience with neutron soil-moisture meters, the instrument has assumed a permanent place among the tools of hydrologists and will continue to be used. The extent to which the method proves useful may depend upon improvements that yet need to be made. There appears to be sufficient need to justify further research into methods of improving the resolution of the instrument, although gains in this direction may be difficult to achieve.

There also appears to be a real need for further developmental work on the instrument itself. The state of the art in electronic and related engineering is such that there seem to be no fundamental barriers to the improvement of presently available instruments. As large projects become more dependent upon the method, equipment breakdown presents an increasing hazard with respect to loss of vital data. Furthermore, if the instrument is to be used to any extent in less developed areas of the world, increased reliability is essential.

More studies seem to be needed on the field calibration problem, especially in relation to heterogeneous soils. Although not a single worker mentioned it, the bulk-density measurement necessary to relate the gravimetric to the volumetric water content is not easily obtained in the field for many soil profiles.

After careful review of the responses on the questionnaires, it is the opinion of the author of this report that few if any of the respondents are in a position to carry out the necessary research on the method except at the expense of other research activities. Those groups with the greatest need for the information appear to have the least time or capability to devote to improvements. Except for the study by Gardner and Roberts,[3] little really current research relating to the method was reported. Appendix C lists those papers relative to development of the method.

Even a casual consideration of the economics of neutron-meter production indicates that the relatively small number sold each year virtually precludes improvements in the system that do not arise out of other projects of the company. However, when one considers the extent of use of the instrument and the financial commitment to the collection of the soil-moisture data in hydrological studies, spending even a modest fraction of this money on improving the instrument and increasing the efficiency of its use would be justified. None of the present research groups will undertake this development work unless funds are made available specifically for that purpose. Conceivably, if this need were brought to the attention of appropriate federal agencies or the IAEA by the IHD Committee, they might be willing to assemble the necessary talent and financial support to undertake the work. Failing this, the IHD should seek means to persuade university or private groups to undertake the development work on a grant or contract basis. The overall budget of such an effort would be small relative to that of many of the IHD projects now being planned, and the potential benefit seems large.

The nature and extent of the response to the questionnaire are greatly appreciated. The response is taken as an indication of a desire on the part of the respondents to see improvements in the method and to receive information on any improvements as rapidly as possible. In view of the work on the method in other countries, most of which is largely unknown to American workers, it is also recommended that the International Work Group on Nuclear Techniques strongly encourage information exchange on this subject by all available means. (FJM)

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APPENDIX A: QUESTIONNAIRE

QUESTIONNAIRE

Please return to W. R. Gardner, Department of Soil Science, University of Wisconsin, Madison, Wisconsin 53706

IHD Questionnaire—Neutron Moisture Meter Use

I. Name of Respondent:_____

Institution:_____

Address:_____

☐ Please place my name on the permanent mailing list.

II. Description of Neutron Meter Used (please use separate form for each different type of meter):

Manufacturer_____ Year purchased_____

Probe: ☐ Surface Electronics: ☐ Scaler

☐ Subsurface ☐ Ratemeter

Source: Isotope(s)_____

Activity_____

Geometry (center or end-mounted source)_____

In this questionnaire the term Meter refers to the complete instrument; the Probe is the sensor head (source, detector, shielding, pre-amp); Surface Probe refers to a sensor used primarily at or near the surface; and Subsurface Probe for a sensor used primarily below the surface.

Please describe the meter in more detail on the reverse side if it is: (a) custom built, (b) an extensively modified commercial meter, (c) an outdated commercial meter for which a catalog description is no longer available.

List any other sensors combined with neutron probe (e.g., gamma ray)_____

_____

III. Calibration: The precision of the results obtainable by the neutron moisture meter depends heavily upon the adequacy of the calibration. As many details of the calibration procedures as you can give will be appreciated. If the procedure is described in the literature, you may substitute the appropriate reference(s) instead of completing this section. If you have a reprint available, this will be appreciated. If manufacturer's calibration only was used, please check ☐

A. Laboratory Calibration:

1. Test container (dimensions, material, etc.)_____

_____

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IHD Neutron Meter Questionnaire (Continued)

2. Composition of porous medium–water mixture (sand, soil, hydrogenous material)_____

_____

3. How was the percent water varied and determined?_____

_____

_____

4. Description of access tube_____

5. Accuracy and precision of measurement (state how defined)_____

_____

At low water contents_____

Intermediate water contents_____

High water contents_____

6. Could you supply a sample calibration curve on request? ☐ Yes ☐ No

B. Field Calibration: Describe procedures used to give accurate measurements in the field for different soil types and aquifer types. Consider both absolute and relative calibration.

1. Reference standard: Absolute reference_____

Day-to-day (e.g., shield count)_____

2. Soil or aquifer characteristics posing special calibration problems, such as chemical composition, structure, nonuniform water distribution, etc.:

3. Comparison with nonnuclear sensors (e.g., gravimetric sampling, resistance units. . .):

Method used_____

Number and distribution of measurements_____

4. Reproducibility or precision of measurements:

Low water contents_____

Intermediate water contents_____

High water contents_____

Comment on limiting factors in reproducibility_____

C. Calibration Research: Briefly describe any special research investigations into effect of soil composition, variation of materials or dimensions of access tubes, temperature, inhomogeneity of soils, etc., upon calibration. Or, preferably, give citation or submit reprint of published description of such research.

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IHD Neutron Meter Questionnaire (Continued)

IV. Access Tubes:

Material used _____

Dimensions _____

Method of installation in field _____

_____

Normal length of useful life in field _____

Special problems or comments _____

_____

V. Field Application: Describe principal applications instrument in hydrological investigations (infiltration, deep percolation, evapotranspiration, saturated porosity, irrigation control, etc. . .). (Reprints of or references to published work would be appreciated.)

VI. Extent of Use:

How long have you used this method? _____

How recently? _____

How often do you use it?

☐ Continuously

☐ Frequently

☐ Occasionally

VII. Overall Evaluation of Method:

1. Accuracy: satisfactory ☐, unsatisfactory ☐
2. Reliability: satisfactory ☐, unsatisfactory ☐
3. Ease of maintenance: satisfactory ☐, unsatisfactory ☐
4. Frequency and availability of repair: a serious problem ☐, minor problem ☐, no problem ☐
5. Comments: _____

_____

_____

VIII. Developmental Needs: State the principal development that you would like to see improve the application of neutron moisture meters to your problem, either in the instrument itself or in the theory.

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Survey of Applications for ^{252}Cf*

By J. L. Crandall†

Abstract: *One gram of ^{252}Cf emits 2.34 × 10^{12} n/sec from a volume of less than 1 cm^3 with a heat production of less than 40 watts and a half-life (alpha + spontaneous fission) of 2.6 years. Because of its high average neutron yield per radioactive disintegration, it produces neutrons less expensively than any comparable radioisotope source and competitively with small nuclear reactors and accelerators. It thus has major potential in providing compact, portable, and reproducible neutron sources at intensities ranging from a few up to 10^{14} n/sec. Such sources have unique applications as tissue implants for cancer therapy, as point sources for neutron radiography, and as portable tools for mineral exploration and also fill more specialized needs, e.g., for making reactor physics measurements. Other uses are expected in such fields as activation analysis, short-lived radioisotope production, moisture gages, well logging, and in-line process instrumentation, where the operating simplicity, minimal maintenance, and fully predictable output of these sources should open many applications currently closed to the more conventional accelerator and reactor sources. Particular emphasis is being placed on industrial uses of the ^{252}Cf sources for in-line process control. A preliminary market survey by Savannah River of possible ^{252}Cf applications indicates a demand for 20 to 60 g of ^{252}Cf by 1975 with the probability of an exponentially increasing demand for some time thereafter.*

Californium has a number of properties[1] that may be profitably applied. Its alpha half-life is 2.6 years,[2] and the average energy is 6.11 Mev per alpha particle.[3] It is thus one of the more favorable isotopic heat sources, having a specific power rating of 39 watts/g (19 watts alpha; 20 watts spontaneous fission). Californium-252's sister isotopes—^{249}Cf and ^{251}Cf—also are attractive for their long half-lives, 360 and 1500 years, respectively,[4] and provide relatively radiation-free starting materials for chemical studies and the production of still heavier nuclides.[5] However, the heat-source users are not going to pay thousands of dollars per watt for their heat sources, nor are the radiochemists going to use kilograms of californium in their experiments. Large-scale uses of californium are possible, instead, because of the spontaneous-fission half-lives[2] of 85.5 years for ^{252}Cf and 62 days for ^{254}Cf, and more particularly because of the attendant neutron production, 3.8 n/fission.[6]

COMPARISON WITH OTHER SOURCES[7-9]

Other Radionuclides

Californium-252 is the lightest nuclide to exhibit a half-life of less than 10^3 years for spontaneous fission.[10] Therefore californium has no competition as a spontaneous-fission source. Since spontaneous fission is a much more efficient neutron producer than (α,n) and (γ,n) reactions, californium surpasses all competing radioisotopic neutron sources in intensity, small size, low heat production, and low helium releases (Table 1).

These same advantages also translate into source prices. Because ^{252}Cf emits several hundred times as many neutrons per curie as its competitors, it can actually be much less expensive on a per neutron basis than other radionuclides costing much less per gram.[7] Further, for large sources emitting 10^{10} n/sec or more, probably only the Sb-Be source, now commercially available from AECL as their Neutroncell system,[11] is even a feasible competitor to ^{252}Cf. The replenishment and shipping costs for the 60-day ^{124}Sb make it both inconvenient and expensive as compared with ^{252}Cf. The potential market for radioisotope neutron sources is large, with some 1000 groups already licensed to use

Table 1 Radioisotopic Neutron Sources

Source	Half-life, years	Yield n/1000 dis	Yield n/μw	α/n
(α,n) sources				
^{210}Po-Be	0.378	0.068	78	15,000
Ra-Be	1,620	0.41	85	2,400
^{238}Pu-Be	89	0.076	91	13,000
^{239}Pu-Be	24,360	0.049	62	20,000
^{241}Am-Be	458	0.054	67	19,000
^{242}Cm-Be	0.346	0.11	83	9,100
^{244}Cm-Be	17.6	0.081	83	12,000
(γ,n) sources				
^{124}Sb-Be	0.167	0.043	2,500	No α
Spontaneous-fission sources				
^{252}Cf	2.65	120	63,000	8.3

*Du Pont Report DP-MS-68-66, presented at Californium-252 Symposium (CONF-681032), Oct. 22, 1968, New York, N. Y., sponsored by the New York Metropolitan Section of the American Nuclear Society. This updated version reprinted by permission of the author and of the proceedings editor.

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Sb–Be, Pu–Be, and Am–Be sources in the United States alone.[12]

Reactors

Since ^{252}Cf is produced in nuclear reactors at a cost, after all the side chains are included, of several hundred neutrons per atom of ^{252}Cf formed, it does not seem that, in returning 0.12 n/atom disintegrating, ^{252}Cf can compete very effectively with a reactor as a neutron source. This supposition is true for the large production and power reactors. However, as the reactor output shrinks, the fixed costs both for the reactor structure and for operations, safety, administration, etc., become disproportionately high, while the utilization of the reactor neutrons often drops precipitously.

A possible compromise between the reactors and the simple ^{252}Cf sources is the use of multiplying sources. When neutron fluxes of about 10^{10} to 10^{12} n/(cm^2)(sec) are wanted, these multiplying sources might be able to achieve some of the low fueling costs of reactor neutron production without incurring the safety and administrative overheads of a true reactor. One version of this device depends on the fact—proved in real "old school" tries at Oak Ridge,[13] AB Atomenergi,[14] and elsewhere[15]—that a natural-uranium, H_2O system can be brought to $k \simeq 1$, but never to actual criticality. Alternatively, it is possible to achieve effectively higher fluxes with a subcritical mass of enriched uranium in H_2O. The Russians have actually used a portable, multiplied Po–Be source for petroleum exploration.[16]

For the reactor–^{252}Cf comparison, it is obvious that, when the work can be brought to a big reactor, the reactor will be favored over californium, but that, when it is a case of moving the mountain to Mohammed, californium can usually win by qualifying as a molehill rather than as a mountain. A rather extreme example of this approach is Oak Ridge's suggestion of using a 20- to 60-g californium source instead of a reactor to study in situ neutron effects in civil defense evaluations.

Accelerators

Californium-252 vs. accelerator comparisons are the most interesting because competition is close in many areas. This is also the area where I can get into real trouble with my accelerator friends by comparing 1980 californium with 1968 accelerators.

As demonstrated in the Canadian studies on their Intense Neutron Generator (ING),[17] large, expensive accelerators have the same cost and neutron intensity advantages as large, expensive reactors so long as the samples can conveniently be brought to the machine and the full output of the machine is used in productive programs. The same thing holds true in lesser degree for medium-sized accelerators such as Linacs,[18] but even these machines are not for every garage. In the smaller sizes, there are Van de Graaffs, cyclotrons, Cockcroft-Walton machines, etc.,[19,20] yielding 10^8 to 10^{12} n/sec, the most attractive range for the californium sources.

The Cockcroft-Walton machines, which produce 14-Mev neutrons by accelerating 100-kev deuterons onto tritium targets, are the most useful. With price tags of $10 to $30 thousand, these machines have always struck me as one of the few genuine bargains in scientific equipment. They can produce neutrons at costs comparable to that of ^{252}Cf, but currently they suffer from the major disadvantage of short target life, which effectively increases their costs and decreases their utility. It has, for example, been a major advance to extend the accelerator target life[21] to 4 days at 10^{11} n/sec. This disadvantage can very probably be overcome in the 1980 machines (although still only to the point of target lives of weeks rather than the years available from californium). The likelihood of breakdowns, the need for regular maintenance and overhauls, and the inherent variability of the neutron intensity will probably always remain with the accelerators. Thus it can be presumed that the small accelerators will continue to be used where their specific characteristics, such as nearly monochromatic 14-Mev neutron production and on–off and pulsed operation, are required; but californium sources will be used whenever ease and reliability of operation and reproducibility of output are primary prerequisites. Californium of course gets the nod in almost all unattended operations.

NEUTRON SPECTRUMS

I have tacitly assumed that the isotopic neutron sources, accelerators, and reactors compete only on cost, intensity, and ease of access. However, other parameters are important. One already hinted at is the neutron-energy spectrums (Fig. 1). The very large accelerators are capable, if desired, of producing very-high-energy neutrons, which are most intriguing but somewhat out of the range of the present study.[22] The D–T accelerators and the ^{124}Sb–Be systems are very nearly line sources, producing 14- and 0.025-Mev neutrons, respectively. The (α,n) sources produce very

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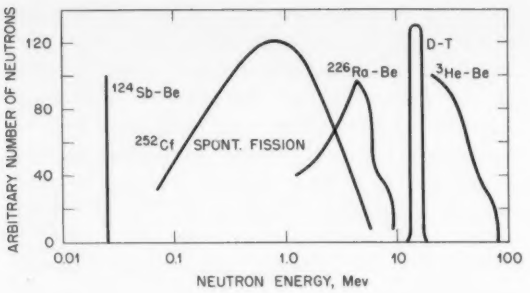

Fig. 1 Energy spectrums of neutrons from different sources.

broad and rather variable spectrums normally centered about 5 Mev.[9] The fission neutron spectrums from both reactors and ^{252}Cf follow a nearly Maxwellian distribution[1,20,23] centered about 2 Mev. A pure fission spectrum is more easily accessible from ^{252}Cf than from a reactor. Moderated spectrums typically show a thermal equilibrium peak, normally around 0.025 ev, connected to the fission neutron spectrum by a slowing-down region. All three spectrum regions have important applications. One of the interesting possibilities for ^{252}Cf (Fig. 2) is being able to moderate only to a rather pure slowing-down spectrum. Another possibility more easily accomplished with ^{252}Cf than a reactor is being able to change the moderator temperature and hence the neutron temperature over a wide range. Very cold (<20°K) neutrons can be used as sensitive probes for measuring lattice phonon distributions, as very-high-cross-section projectiles for activation reactions, and in neutron radiography.[24]

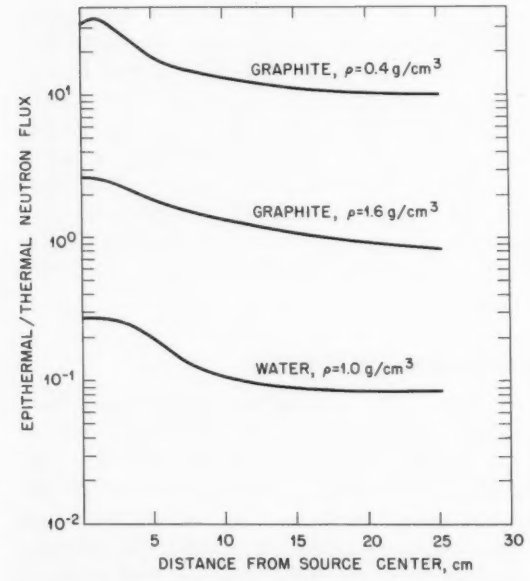

Fig. 2 Epithermal (0.025 ev to 9.12 kev) to thermal (0.0 to 0.625 ev) flux ratios for moderated ^{252}Cf sources. (Calculated by Hammer and Anisn codes for ^{252}Cf source at center of moderator sphere.)

SHIELDING AND SAFETY

Extending as they do over an energy range of 10^{10} or better, the different neutron spectrums can lead to different neutron uses. They also (Fig. 3) have quite different shielding requirements.[19,20,25] The advantage in reduced shielding which ^{252}Cf enjoys over the D-T accelerators because of lower neutron energies is augmented by a second shielding advantage due to smaller volume. The mass of shielding required varies between the first and second power of the source volume. For reactors and many accelerators,[26] a whole room is usually shielded rather than a few cubic centimeters as for ^{252}Cf. However, it should be emphasized that even for ^{252}Cf the shielding requirements are not trivial. The <1 g weight of a 10^{11}-n/sec ^{252}Cf source becomes a weight of 1 to 2 tons with a corresponding radical decrease in the source portability by the time it is shielded to tolerance levels.

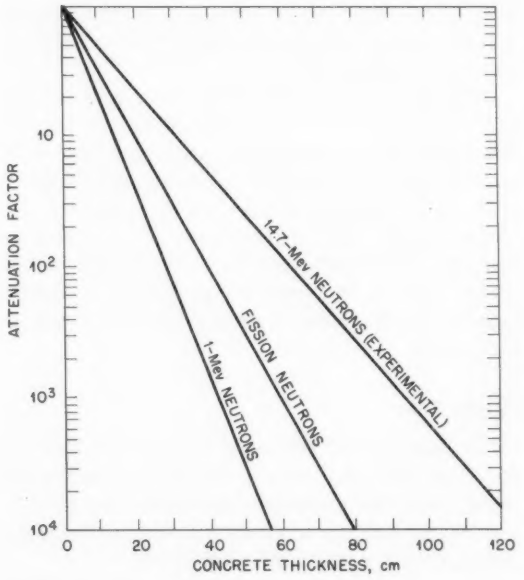

Fig. 3 Neutron shielding requirements (from Ref. 20).

A comparison of the safety aspects of the different neutron sources shows that reactors are the most hazardous:

Reactors	Accelerators	Radioisotopes
Primary hazards		
Nuclear excursion	Radiation exposure	Radiation exposure
Fission-product release	Electrical shock	Radioisotope release
Radiation exposure	Tritium ingestion	
Potential seriousness		
Large	Small	Intermediate
Accident safeguards		
Automatic and manual controls	Shielding	Shielding
Shielding	Interlocks	Radioisotope canning
Containment		
Safety effort required		
Large	Small	Small

Some of the major expenses in the use of reactors stem from the precautions taken to decrease accident likelihood to acceptable (and minuscule) levels.

Radioisotope sources are the next most hazardous because of their easy portability, the fact that they are never turned off, and the extremely low permissible levels for biological uptake of the radioisotopes[27] (0.04 μc maximum permissible body burden for ^{252}Cf).[28] Fortunately these hazards with radioisotopes can be circumvented by good packaging. Multiple containment barriers must be used, particularly in view of the unlovely habit radioisotopes have of spreading themselves from an unsealed environment through fission-product and alpha-emitter recoils.

The hazards from the accelerators are generally small. However, a number of accidents have occurred when people have contacted the operating beam.[29] Accelerators have a higher probability for minor accidents, such as electrical shock or tritium inhalation from mishandled targets.[30]

^{252}Cf NEUTRON USES

The uses of ^{252}Cf in relation to the neutron properties are systematized in Fig. 4. Certainly the first point obvious from this chart is that there are so many uses. The second point is that most of these potential uses, although by no means fully explored, are even now represented by specific applications.

Direct Neutron Counting

Perhaps the least glamorous, but one of the most widespread, uses for ^{252}Cf may be as a standard neutron source for calibrating neutron-detecting instruments. Although presumably slightly less reproducible than the longer-lived Ra-Be, Pu-Be, or Am-Be sources, the ^{252}Cf sources have the advantage that they can be made intense enough to test the often nonlinear instrumentation in the actual range in which the instrument is to be used.[31] The ^{252}Cf sources should also be cheaper than competing radioisotope sources.

Another widespread use for ^{252}Cf sources should be in the startup of nuclear reactors. With a fresh fuel charge in the reactor, a fairly copious artificial source of neutrons must be provided during startup so that the instruments can read the multiplication of this source and hence the distance from criticality[32,33] (Fig. 5). Here again, intensity and cost favor ^{252}Cf over the now standard Po-Be and Sb-Be sources. An extension of this use is using ^{252}Cf sources in subcritical multiplying systems to substitute for nuclear reactors and thus providing cheaper and safer training in reactor use and operation.

A unique but small-scale reactor use for ^{252}Cf is in reactor physics measurements.[34] Essentially all the traditional detailed measurements of reactor physics determine the effects of removing neutrons in activating foils or detecting instruments. The tiny ^{252}Cf sources, since they emit neutrons in the normal fission spectrum of the reactor, offer the possibility of determining the effect of adding neutrons at localized points in the reactor, and hence give access to the much theorized-about but hitherto almost unmeasurable neutron importance function for the reactor.

Californium-252 can also be used in criticality monitoring. The Atomic Energy Commission predicts that by 1980 the power-reactor industry will be burning some 72 thousand kg of ^{235}U a year while producing 29 thousand kg of ^{239}Pu a year.[35] These amounts of fissionable materials will be circulated through feed-chain pipelines many times, and all will carry with them some hazard for accidental criticalities. Several such criticality incidents have occurred, and others will occur in the future unless greatly improved instrumentation is provided to warn of incipient criticality.[36] Unfortunately, really satisfactory instrumentation has proved extremely difficult to design and is still not available even in concept. However, we do know such instrumentation will have to depend on neutron-multiplication measurements of

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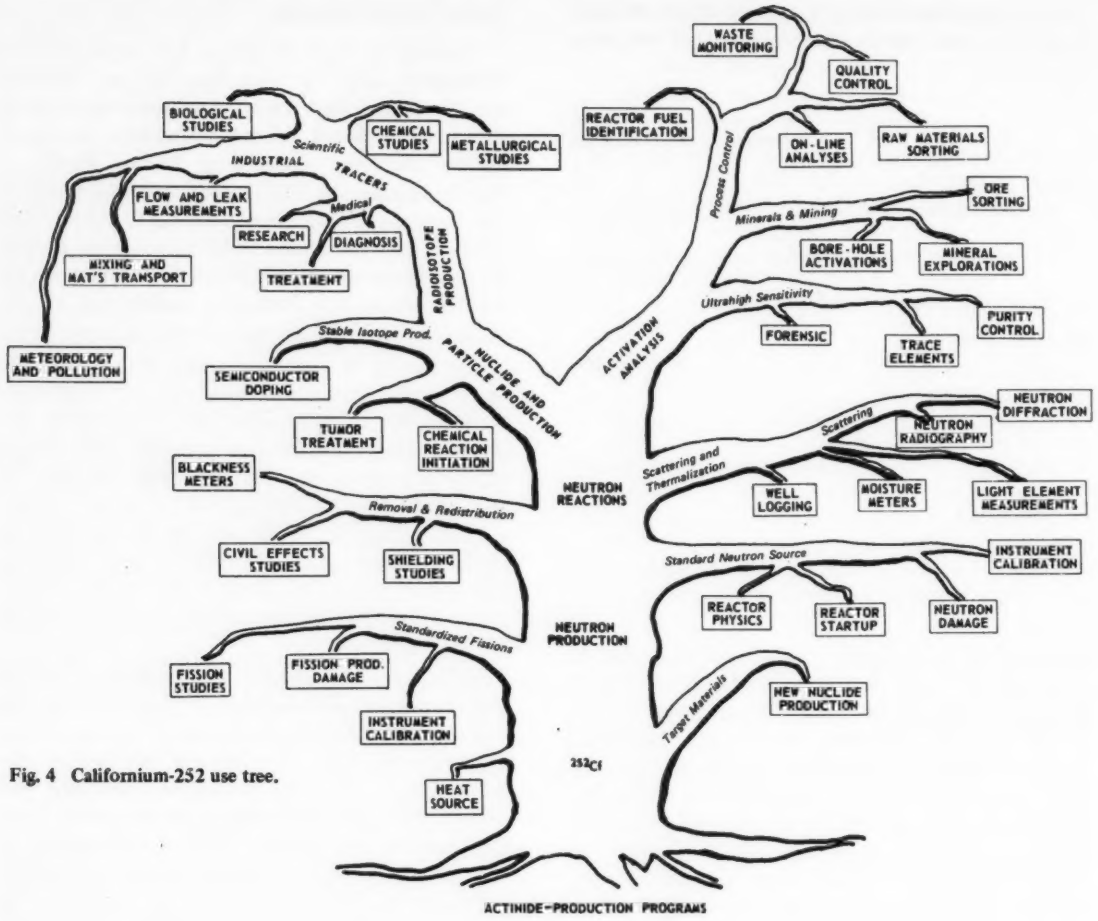

Fig. 4 Californium-252 use tree.

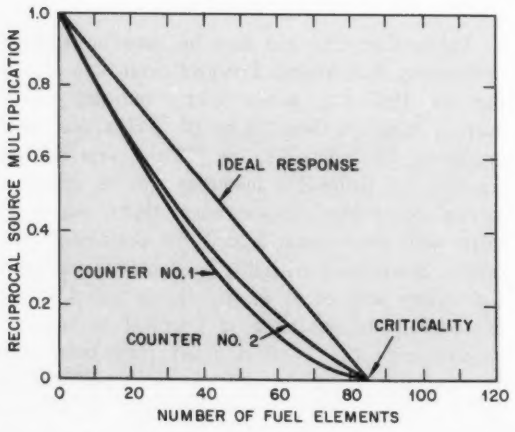

Fig. 5 Source multiplication curves for subcritical assembly.

some sort. Thus here is an eventual major source demand in which, unless the reliance is entirely on some kind of very-short-burst pulsed-neutron methods, ^{252}Cf would be preferred because of its reliability for continuous, unattended operation.

Neutron Scattering

If the simple neutron-source applications provide the least glamorous uses for ^{252}Cf, neutron scattering provides the most glamorous ones. Neutron radiographs[37] make lovely covers for annual reports displaying the onward march of science. They also have some very down-to-earth uses industrially[38] and some high potential in medical and biological applications.[39] Most of these uses stem from the fact that neutrons image the light elements, particularly hydrogen, much

better than X rays. Californium-252 should almost revolutionize this field as a high-intensity point source.

Neutron diffraction is better developed technically than neutron radiography and has greater superiority over the competing X-ray techniques in its particular use areas.[40] However, even with ^{252}Cf sources, neutron diffraction may be limited by neutron intensity/cost considerations. X-ray diffraction equipment is used in every major scientific laboratory and university for two primary uses: "fingerprint" identification of chemical compounds by the diffraction patterns from crystalline powders and atomic-structure determinations by the diffraction patterns from single crystals. Neutron diffraction is unlikely to compete in the first use, but, since most current structure measurements are being made on organic and other hydrogenous compounds, the fact that neutron diffraction locates hydrogen atoms, while X-ray diffraction does not, would, other things being equal, make neutron diffraction the method of choice for structure studies.[41] Neutron diffraction also has the advantage that it can determine the magnetic structure and phonon spectrum of a crystal lattice whereas X-ray diffraction cannot.

The difficulty with neutron diffraction is that current techniques require well-collimated, monochromatic beams of thermal neutrons. These are obtained very inefficiently from the isotopic, polychromatic neutrons typical of moderated fission spectrums. A thermal flux of 10^{13} n/(cm^2)(sec) is near minimal for extracting a neutron diffraction beam, and obtaining such a flux requires about 100 g of ^{252}Cf. This amount might, at most, be cut by a factor of 10 by going to some sort of neutron howitzer geometry that concentrates the neutrons in the direction of the beam. A better hope is that the divergent beam techniques now being applied to X rays can be adapted to neutron diffraction to take advantage of the intrinsic point geometry of californium sources.[42]

Neutron Thermalization

Many possible uses of ^{252}Cf sources depend on thermalization and subsequent back reflection of fast neutrons by hydrogen and other light elements. The most advanced of these techniques is well logging,[43,44] which is very extensively used to determine water- and oil-bearing layers as well as other stratigraphic information. Actually neutron well logging can involve a number of activation techniques (Fig. 6).[43] Complementary techniques have been worked out using isotopic sources for continuous measurements and miniaturized accelerator sources for pulsed measurements. These techniques are yielding valuable results, and it is expected that their applications and value will increase greatly as the present empirical methods are buttressed with a greater theoretical development.

Neutron moisture gages for other less-specialized environments are well developed and commercially available.[45,46] While nothing could be more ubiquitous than the need for moisture measurements in agricultural and industrial processes, there is a wide variety of related applications, most of which have received only cursory attention. Examples are gaging and quality control of plastics and other hydrogenous industrial materials and the sorting of meat, fat, other foods, and biological materials. The above uses are perforce now being exploited with conventional radioisotopic sources and to a lesser degree with accelerators. For example, more than 6 hundred Pu-Be, Am-Be, Ac-Be, and Ra-Be sources are in use for well logging. If the source comparisons given in the first part of this article hold up, many of the present sources would be desirably replaced by ^{252}Cf sources. The availability of much higher intensities and smaller dimensions for the ^{252}Cf sources should result in many new applications.

Neutron Shielding Studies

A rather restricted use for neutron sources, but one which may require very large source sizes because of the large attenuation factors, is neutron shielding and civil defense measurements. The driving impetus for shielding measurements is the multimillion-dollar savings to be made by eliminating unnecessarily conservative restrictions from reactor, process-equipment, and shipping-cask designs in the power-reactor industry.[47] In the civil defense measurements, neutron transmission, reflection, and capture must be determined in the complicated natural environment.[48] Extreme difficulty has also been encountered in calculating neutron transmission through ducts and labyrinths.[49] Because of their high intensity, small volume, and low heat production, ^{252}Cf sources are overwhelmingly favored over their competitors in these studies.

Neutron Damage Studies

The energy deposition which neutrons make in shielding is also of interest in a number of other contexts. Neutron damage to reactor materials has recently been pushed into the forefront by the Oak Ridge discovery of severe aluminum embrittlement in the High Flux Isotope Reactor (HFIR) and the relation of this embrittlement to fission neutron fluences of 10^{21} nvt and above.[50] Since similar or higher dosages are predicted for the core materials in

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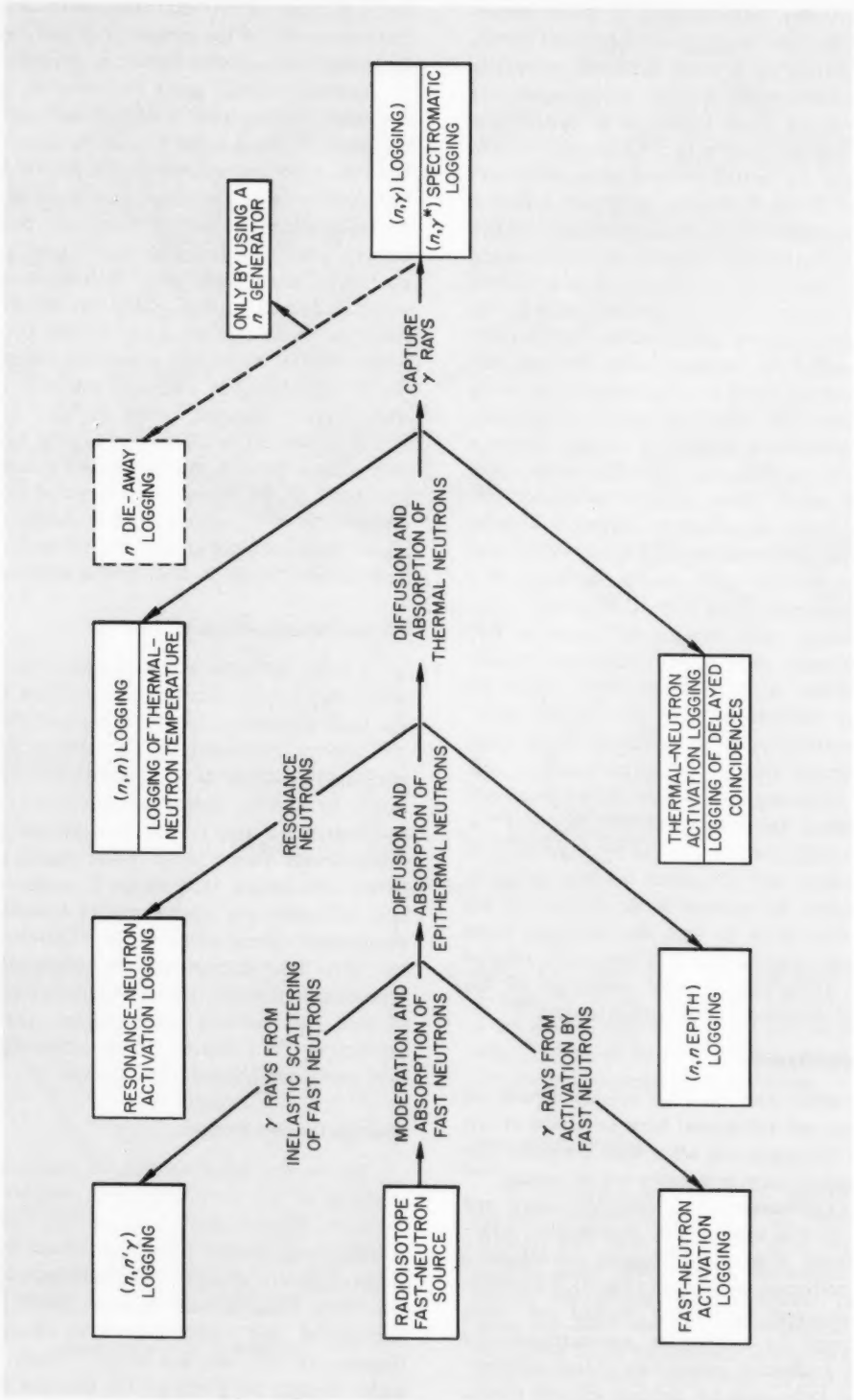
(n,n') LOGGING
RADIOISOTOPE FAST-NEUTRON SOURCE
γ RAYS FROM INELASTIC SCATTERING OF FAST NEUTRONS
RESONANCE-NEUTRON ACTIVATION LOGGING
MODERATION AND ABSORPTION OF FAST NEUTRONS
γ RAYS FROM ACTIVATION BY FAST NEUTRONS
FAST-NEUTRON ACTIVATION LOGGING
(n,n) LOGGING
LOGGING OF THERMAL-NEUTRON TEMPERATURE
RESONANCE NEUTRONS
DIFFUSION AND ABSORPTION OF EPITHERMAL NEUTRONS
(n,n EPITH) LOGGING
n DIE-AWAY LOGGING
ONLY BY USING A n GENERATOR
DIFFUSION AND ABSORPTION OF THERMAL NEUTRONS
THERMAL-NEUTRON ACTIVATION LOGGING
LOGGING OF DELAYED COINCIDENCES
CAPTURE γ RAYS
(n,γ) LOGGING
(n,γ*) SPECTROMATIC LOGGING

Fig. 6 Possible neutron-logging techniques using radioisotopic neutron sources (from Ref. 43).

the breeder reactors,[51] such effects are going to require long-term study. Californium-252 sources are ideal for these studies because they can duplicate reactor conditions so far as neutron fluxes and spectrums are concerned and give much better control over dosages and irradiation conditions.

It is also hoped that the radiation damage produced by ^{252}Cf neutrons can be used beneficially in cancer treatment. The ^{252}Cf sources open up new horizons because they can be implanted at the tumor site. Implantation eliminates the major stumbling block which has always been associated with neutron beams, that of damaging the skin and intermediate tissue in penetrating to the tumor.[52]

Neutron Removal

The remaining ^{252}Cf applications all depend on the capture of spontaneous-fission neutrons in target nuclei.

Neutron removal by capture is obviously the ultimate arbiter in the shielding and civil defense studies discussed earlier. It also finds limited use as an analytical tool. "Blackness testers," which measure the neutron loss in traversing an absorbing sample, have long been used in testing nuclear reactor fuel and control components; there is no reason why these testers could not be applied to any system where the concentration of a neutron-absorbing element in a relatively nonabsorbing matrix must be known. While these applications will always be special cases, the wide spread in cross sections between the different elements guarantees that appropriate test situations will arise fairly often. A further elaboration on this idea is to surround the neutron source with a subcritical multiplying medium that increases the sensitivity to neutron removal (or addition, in the case of a fissioning sample). Again, these devices are used regularly to test reactor components.[53,54] Californium-252 would normally be preferred to the Ra-Be, Pu-Be, and Po-Be sources now used (Fig. 7).

Stable-Nuclide Production

The production of stable and relatively long-lived nuclides by neutron capture has been almost the

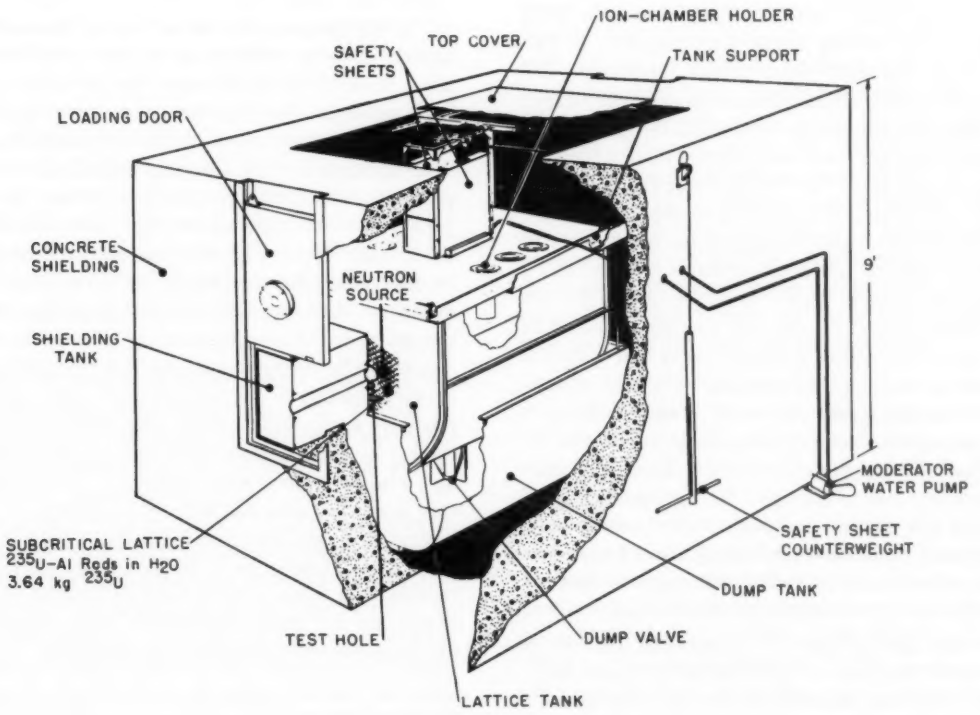

Fig. 7 Neutron-source-testing system.

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exclusive province of nuclear reactors, and ^{252}Cf sources will never compete with reactors in any normal production process. However, the exception might be in "doping" reactions, where it is desired to add parts per billion of some crucial trace element by transmuting atoms at selected locations in situ. Such processes are in very limited use in commercial manufacture of transistor and integrated circuits.[55] The convenience of manufacturing an article entirely in one's own plant with a ^{252}Cf source would normally outweigh the lower neutron costs of irradiating at a large reactor. The eventual utility of transmutation doping is hard to judge, but there is no doubt that parts-per-billion impurities are becoming industrially important.

Short-Lived-Radionuclide Production[56,57]

Californium-252 sources are much more likely to play an important role in transmutations to the very-short-lived nuclides than to the very-long-lived ones. The short-lived nuclides have one very marked advantage over the normal medium-lived nuclides for tracer and irradiation studies—they permit working with relatively high levels of radiation during the study period without leaving a long-term radiation residue. For several elements—e.g., oxygen, aluminum, and silicon—only short-lived isotopes are available. Despite these potential advantages, the logistic difficulties with the short-lived nuclides have severely limited their use except in cases like $^{113}Sn/^{133m}In$, where the short-lived nuclide can be eluted from a longer-lived generator.[58] Californium-252 plus an appropriate target material affords a sort of universal generator, which can, if desired, be coupled to the test environment for the short-lived nuclide. Indeed, the test nuclide could be generated from existing materials in the test apparatus by passing neutrons through the walls without ever directly disturbing the test setup. Tracer techniques could become routine for testing industrial and laboratory operations in place and even for such mundane purposes as leak detection. In the corresponding biological and medical uses of the short-lived isotopes, the emphasis would be on decreasing the radiation dose to the patient in tracer studies and in providing more precisely controlled dosages in radiation treatment.

A very special use for in situ production of short-lived nuclides is with activable-tracer techniques.[59] Here one is permitted the luxurious paradox of using a nonradioactive tracer for radioactive counting by adding a stable, but normally scarce, isotope to, say, a biological system and then detecting the isotope migration path by after-the-fact irradiation.

Autoradiographs of activated specimens belong in the same category. This technique has incidentally been carried full circle by activating the silver salts in faded photographs to recapture the image as an autoradiograph of the original.[60]

The definition of a short-lived nuclide depends to some extent on the location of the user. In the United States it is about 2 hr, which is about the shortest time needed to fly the nuclide from the producer and to get it ready for use.[61] In one of the lesser-developed countries, the definition might extend out to half-lives of many days because of the differing cost and convenience factors, and indeed the same thing might apply in the United States for repeated rather than one-shot uses. Thus there is a considerable range of applicability. Short-lived isotope production could well be one of the chief uses of ^{252}Cf, particularly since it could in part be relegated to off-hours when the source would otherwise be yielding unproductive neutrons.

Localized Energy Deposition

In the extreme case the half-life of the short-lived nuclide drops to milliseconds or less, and the irradiations have to be made in place. A rather limited application of this situation is in providing highly localized and very heavy energy depositions from the decaying nuclide.[62] A fissioning nuclide, for example, provides about 160 Mev of kinetic energy to fission products, which then travel only a few microns in a solid. The $^{10}B(n,\alpha)^{7}Li$ reaction, although less spectacular, still liberates about 2.5 Mev over similar distances. Such massively localized energy releases, besides having interesting research applications, should also be useful for catalyzing chemical reactions and for tumor destruction. In fact, several years ago some relatively unsuccessful efforts were made to treat brain tumors by impregnating them with boron compounds and irradiating them with neutrons.[63]* The failures of this technique were ascribed to an inability to find a boron compound that would localize in the tumor and to excessive damage produced by the neutron beams in reaching the tumors. Both these difficulties could be minimized by using implantable ^{252}Cf sources and localizing the neutrons rather than the boron.

*A very recent news item from Japan reports a considerable improvement in this technique.

Activation Analysis[20,64-68]

The remaining and perhaps the most important class of uses to be discussed for ^{252}Cf neutrons is activation analysis. Although still rather more discussed than performed, activation analysis is a well established field with an extensive literature, proven techniques, and full-time practitioners. It also has the advantage of lying along several main lines of development. On the one hand, increasing automation and increasing strictness of quality control are creating increasing demands for in-line nondestructive analyses and for ultrasensitive analyses, both of which can be provided by activation techniques. On the other hand, such parallel advances as high-resolution lithium-drifted germanium detectors[69] (and hopefully their room-temperature successors[70]) and on-line computer data-taking and analysis, not to speak of ^{252}Cf sources, are rapidly adding to the activation-analysis capabilities.

The ultrasensitive activation analyses are very impressive and have served such spectacular ends as finding arsenic in Napoleon's whiskers[71] and detecting 2000-year-old counterfeiting of Greek coins.[72] However, this sort of study will always be performed with nuclear reactors or the superaccelerators. The sensitivity for most analyses is directly proportional to the flux level, and even fairly ordinary research reactors now offer thermal-neutron flux levels in the 10^{14} n/(cm^2)(sec) range as against a maximum of about 10^{12} n/(cm^2)(sec) even for a 1-g californium source with subcritical multiplication. However, if one is willing to accept sensitivities of micrograms or even milligrams[73] rather than nanograms,[74] either the ^{252}Cf sources or the smaller accelerators producing 10^{10} to 10^{12} n/(cm^2)(sec) provide analytical results directly at the using laboratory rather than at a remote reactor installation.

The high-sensitivity measurements are often quite time-consuming, requiring as much as a week of irradiation time, and sometimes involving both pre- and postirradiation chemical separations, as well as complicated counting techniques to avoid interferences. For in-line process use of activation analysis, however, normally only percentage compositions are needed, so that sensitivity may be traded for time.[75,76] Also, either the application is carefully selected to avoid complications, or instrumental and computer methods are used to resolve the interferences. In most respects, however, activation analysis is less restrictive than other methods of analysis. For instance, it is one of the few nondestructive techniques that can be used for nondestructive analyses of powders, slurries, and bulk solids.

As with conventional activation analysis, neutrons of thermal, resonance, or fast energies may be used to enhance sensitivities for certain elements and to avoid interferences with others in the in-line applications. Half-lives of the activation products should ordinarily be short, both to maximize the number of counts available from a short irradiation and to decrease time delays in feeding back the analytical data into the process controls. A tempting possibility is prompt-gamma measurements on the neutron captures or inelastic scatterings.[77,78] In these reactions, which often produce extremely high-energy gamma rays, essentially all the decay radiation from the compound nucleus is made available to the detector within 10^{-12} sec. Isenhour and Morrison[77] have calculated that, at equal activating flux levels, prompt-gamma measurements should give better sensitivities than delayed-gamma measurements. However, there are problems in separating the source gammas from the activation gammas.

Delayed-gamma measurements are of particular interest in flowing process systems. For example, where the rate of flow and branching of specific materials are crucial, a simple activation at one point with subsequent detector measurements in the different flow streams can solve the problems.[79] Note that this technique will even work for two different materials of approximately the same elemental composition because it is only necessary to activate one of the materials in its original stream. In the opposite situation where flow conditions, or at least rates of movement, are fairly stable, as with a conveyor belt, detectors at different distances downstream from the activating source provide an automatic method for time sorting of different half-life activators.[75]

For in-line process activation analysis, reactors are out of the picture, and semiportable sources emitting 10^{10} n/sec or better are required. Only the D-T accelerators and ^{252}Cf are likely to qualify, and ^{252}Cf has the advantage of lower shielding requirements, much more uniform neutron output, and complete absence of maintenance requirements. However, the 14-Mev neutrons from the accelerators are able to produce the reactions for determination of three very crucial light elements—$^{16}O(n,p)^{16}N$, $^{14}N(n,2n)^{13}N$, and $^{28}Si(n,p)^{28}Al$—better than the fission spectrum from ^{252}Cf sources.[80] For these particular reactions the ^{252}Cf source would have to be an order of magnitude stronger than the accelerator source in order to yield

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equal sensitivity. This would also be the case for many of the inelastic scattering measurements, but here scattering and capture measurements are often competing and the seeming disadvantages of the ^{252}Cf may be an actual advantage.[81] The major advantage of the ^{252}Cf sources is in the calibration irradiations needed to put the unknown activation samples on a quantitative basis. With the accelerator, a standardizing irradiation must be performed with each unknown, and this requires complicated double rotating rigs to get uniform exposures.[82] With the highly reproducible ^{252}Cf sources, such standardizing irradiations can be reduced in numbers, repeated if need be, and performed before and after the fact, as well as simultaneously with the test irradiation. The conclusion seems to be that ^{252}Cf will be favored for the in-line applications except where specific analyses for oxygen and nitrogen are required. A very interesting paper on exploratory tests of ^{252}Cf for activation analysis has been presented by Keller, Ferguson, and Nichols from ORNL.[83]

CALIFORNIUM DEMAND PROJECTION

In conclusion, I am presenting the results of a survey of possible ^{252}Cf use demands made by Reinig[84] at the Savannah River Laboratory (see the tabulation below).

This survey goes only through calendar year 1975 and thus really covers only development demands, but it indicates requirements of 20 to 60 g of ^{252}Cf, enough to justify at least the initial phase of the production programs now being considered. (MG)

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Reference marked with an asterisk has not been verified by the editors; for further explanation, see the Foreword.

An effort was made to provide a follow-up reference for each subject discussed. However, no attempt was made to provide a comprehensive bibliography.

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Use	Amount, mg							Total, g
	1969	1970	1971	1972	1973	1974	1975	
Radiotherapy	0.4	.0.4	1	10	20	30	40	0.1
Radiography	5	100	200	600	1000	2000	2000	6
Neutron activation analysis	10	60	100	400	500	500	500	2
Hydrogen and petroleum exploration	5	10	30	50	200	400	400	1
Nuclear safeguards	10	50	300	700	1000	3000	5000	10
Industrial applications	5	10	50	100	400	500	1000	2
Research and training	1	10	50	100	200	300	400	1
Civil effects, g	–	–	–	20–40	–	–	–	20–40
Total, g	0.036	0.240	0.731	1.96 22–42	3.32	6.73	9.34	22 42–62

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Instrumental Analysis of Neutron-Irradiated Soils*

By J. R. Kline and S. S. Brart

Abstract: *Neutron-activated soils from a worldwide collection were analyzed by gamma-ray spectrometry to identify and measure trace elements that can be studied by this technique and to determine whether gamma-ray spectra of soils can be used to identify specimens of soil and trace them to their geographical origin. Samples of 100-mg size were irradiated for either 5 or 10 min in the Argonne CP-5 reactor at a thermal-neutron flux of 2×10^{13} n/(cm^2)(sec).*

Nuclides with half-lives shorter than 15 hr were not measured. Long-lived nuclides, which could be observed after 4 days' delay, are ^{140}La, ^{46}Sc, ^{82}Br, ^{59}Fe, ^{51}Cr, ^{60}Co, ^{152}Eu, ^{154}Eu, ^{153}Sm, ^{181}Hf, and ^{95}Zr–^{95}Nb. Quantitative nondestructive determinations were easily made for La, Sc, Fe, Co, Eu, and Sm. Amounts of scandium were correlated with amounts of iron, cobalt, and europium in soils of highly diverse origin. Similar correlations were not obtained with lanthanum. Elemental relationships sometimes resulted in similar gamma-ray spectra for soils of dissimilar origin. These spectra, therefore, raised doubts as to whether they could be used without other supporting evidence to identify soil specimens or trace them to their place of origin when the analysis is done with NaI crystals and 400-channel analyzers.

Neutron activation, when combined with gamma-ray spectrometry, is a potentially useful analytical technique in soils investigations. It may be used in conjunction with destructive radiochemical methods for both the isolation and measurement of practically any element with a suitable neutron-capture cross section and half-life and for which a suitable chemical separation exists. Kline et al.[1,2] used this technique for the fractionation and measurement of copper in several soils; however, methods are frequently inconvenient and tedious. A more attractive method includes the analysis of soils by instrumental methods involving no radiochemistry. Such nondestructive gamma-ray spectrometry may have a more limited range of usefulness, however, because the gamma radioactivity of some nuclides may be obscured by the activity of other nuclides in the spectrum so that these nuclides may go undetected.

This study was made to examine the range of the potential use of nondestructive neutron activation analysis in soils investigations and to identify elements for which this method may be recommended. The operational objectives were: to measure the decay rates of neutron-irradiated soils; to identify at least potentially measurable nuclides in the gamma-ray spectra; to measure some of the nuclides; and to observe the spectral structures of soils and thus determine whether such spectra can be used to identify and match specimens. Matching of specimens by this method has been reported by Bryan, Guinn, and Settle[3] for materials including paint, paper, narcotics, hair, and soils in criminology. This success suggests that the matching procedure could also be useful in soil correlations. Kline et al.[1] noted previously, however, a remarkable similarity among the gamma-ray spectra of soil samples obtained from sites of similar geological parent materials, even when the sites were far apart.

MATERIALS AND METHODS

Analyses were made of soils obtained from a broad spectrum of samples, many of which came from a collection originally gathered during studies of worldwide fallout. Other samples were collected by the authors from southern Minnesota and northern Alabama. The worldwide fallout specimens were finely pulverized and were in a clean, dry condition stored in polyethylene bags that had been inserted in covered cardboard containers. Samples collected by the authors were protected from contamination at all stages of processing and were not pulverized. Table 1 summarizes the origins of soils used in this study.

Details of sample preparation and irradiation were given previously by Kline et al.[1,2] Briefly, this method includes the weighing of 100-mg samples into specially constructed polyethylene packets, which are then heat sealed. From six to eight such packets were placed in a polyethylene capsule and irradiated simultaneously, along with appropriate chemical standards, in the Argonne CP-5 reactor. Irradiations were for either 5 or 10 min in a thermal-neutron flux of 2×10^{13} n/(cm^2)(sec).

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Table 1 Origin of Soils Used in Survey of Neutron Activation Methods in Soils Analysis

Points of origin	
Lebanon	WWF,* lab. No. 601083, Beirut
Malaysia	WWF, lab. No. 611, Singapore
New Zealand	WWF, lab. No. 61117, New Auckland
Hawaii	WWF, lab. No. 601078, Oahu
Norway	WWF, lab. No. 60884, Vadso
Japan	WWF, lab. No. 601086, Tokyo
Brazil	WWF, lab. No. 6113, Belem
Australia	WWF, lab. No. 6120, Alice Springs
Southern Rhodesia	WWF, lab. No. 614, Salisbury
Alabama	AC,† Fullerton silt loam, Jefferson County
Alabama	AC, Linker fine sandy loam, Jefferson County
Minnesota	AC, Fayette silt loam, Wabasha County
Minnesota	AC, Kilkeny clay loam, Steele County

*WWF = worldwide fallout collection.
†AC = authors' collection.

Data collections were initiated 4 days later to allow the decay of short-lived isotopes and were intermittently continued for 62 days.

The counting apparatus was a solid cylindrical NaI(Tl) 13- by 10-cm (5 in. by 4 in.) crystal and photomultiplier assembly, connected to a 400-channel pulse-height analyzer. The energy calibration of the analyzer was maintained at 10 kev/channel by periodic counting of ^{137}Cs and ^{60}Co standards. Samples were counted for 10 min each, and data collections were made over the energy range of 0 to 4 Mev; however, practically all gamma rays of interest were found in the range 0 to 2.1 Mev, and the spectra shown in this report are plotted over this interval. When these data were used for quantitative purposes, corrections were made for background. No such corrections were made, however, when data were plotted for spectral comparisons among samples.

These counting conditions were suitable to measure most isotopes, except ^{152}Eu, ^{154}Eu, and ^{153}Sm. They have low-energy gamma rays and X rays that cannot be resolved effectively on a 0- to 4-Mev energy scale. Accordingly, a very thinly clad 2 by 2 NaI(Tl) crystal was connected to the pulse-height analyzer and calibrated, in turn, on an expanded scale of 2 kev/channel. The low-energy gamma rays and X rays of these isotopes were well resolved and prominent on the expanded scale and hence were suitable for the quantitative measurement of europium and samarium.

RESULTS

Radioactive Decay in Soils

Beginning on the 4th day, and continuing for 62 days thereafter, the samples were first counted daily and, later, weekly or biweekly. The data from each counting run were integrated over the entire spectrum from 0.1 to 2.1 Mev, corrected for background, and plotted against time.

Figure 1 shows representative gross gamma-decay curves obtained from soils of highly diverse origin. These curves were extrapolated to zero time to obtain an estimate of the original radioactivity; because ^{24}Na is known by direct observation to be a dominant nuclide in the spectra, a 15-hr half-life was assumed for the extrapolation. The extrapolations indicate that many of the soils may have initial count rates in excess of 10^8 cpm. Nishita[4] studied both real and computer-simulated curves for soils and found in the latter that omission of the radioactive contribution of nuclides having less than a 3.3-hr half-life reduced the initial radioactivity by almost a factor of 100. This implies, therefore, that many of the soils reported here may have had gross disintegration rates of close to 10^{10} dpm at time of discharge from the reactor. In any case, the rates are obviously much too high to count as the

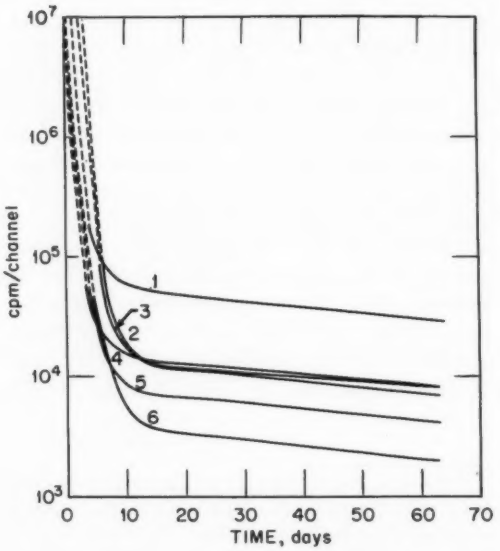

Fig. 1 Gross-gamma-decay curves for six soils. Dashed portion of curve is extrapolated with 15-hr half-life. Curve 1, Hawaii; 2, Malaysia; 3, Minnesota (Fayette); 4, Minnesota (Kilkeny); 5, Lebanon; 6, New Zealand.

samples come from the reactor. Furthermore, nondestructive neutron activation analysis cannot be used to measure nuclides that have half-lives shorter than ^{24}Na, when the irradiation and counting conditions are as specified, since this is the dominant nuclide. From the discussion of total initial count rates, it is apparent that reductions in activity by a factor of about 10^5 would be required to give reasonable final count rates. If this were done by irradiating in a flux of 2×10^8 n/(cm^2)(sec), for example, certain large-cross-section, short-lived nuclides might be measurable, in spite of the ^{24}Na. As a general rule, however, it would seem that, for most elements with "short" half-lives, radiochemical separation or high-resolution counting using Ge(Li) detectors might be more effective methods of measurement.

Gamma-Ray Spectra of Soils

Gamma-ray spectra of the soils were observed at frequent intervals throughout the 62-day experimental period. During the interval from 4 to 12 days, corresponding to the rapidly curving portion of the decay curves (Fig. 1), several isotopes with half-lives up to 40.2 hr could be observed. Beyond this period, the spectra were dominated by two longer-lived isotopes, ^{46}Sc and ^{59}Fe. A list of the elements identified, and some of their physical properties, is presented in Table 2.

Selected spectra for several of the soils, taken 6 days after irradiation, are shown in Fig. 2. Not all soils are included because of the similarity of their spectra, which show, in general, that the same complement of elements were made radioactive in all the soils of this collection. The ratios of these elements are

Table 2 Nuclides Found in Gamma-Ray Spectra of Thermal-Neutron-Irradiated Soils

Nuclides	Half-life,[5] time	Cross section, barns[6]	Abundance in earth's crust,[7] ppm
^{46}Sc	83.9 days	12	5
^{140}La	40.2 hr	8.2	18
^{59}Fe	45.1 days	0.98	5.0%
^{60}Co	5.24 years	20	40
^{152}Eu	12.7 years	1400	1.1
^{153}Sm	1.96 days	140	6.5
^{181}Hf	44.6 days	10	4.5
^{95}Zr	65.0 days	0.09	220
^{24}Na	14.97 hr	0.53	2.8%
^{82}Br	1.5 days	3.1	2.5
^{76}As	1.1 days	5.4	5
^{51}Cr	27.8 days	13.5	200

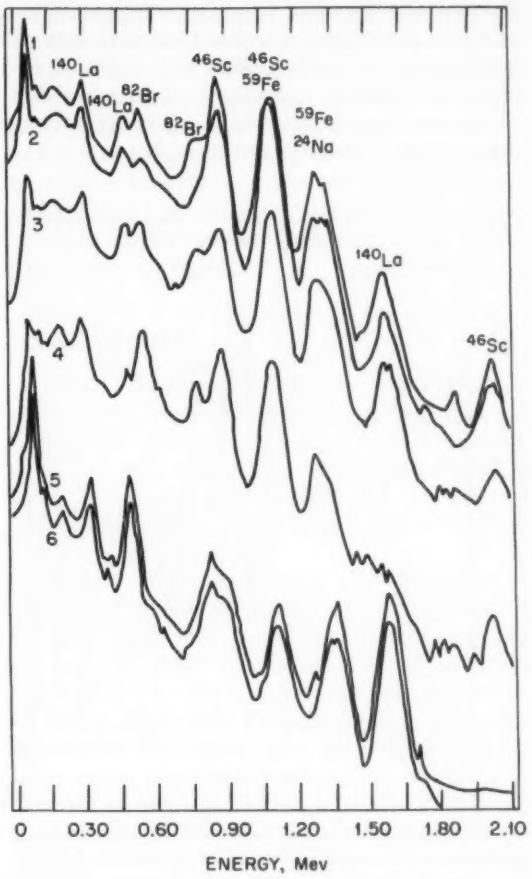

Fig. 2 Gamma-ray spectra of soils taken 120 hr after 5-min irradiation with thermal neutrons at a flux of 2×10^{13} n/(cm^2)(sec). Curve 1, Hawaii; 2, Southern Rhodesia; 3, Minnesota (Fayette); 4, Singapore; 5, Alabama (Linker); 6, Alabama (Fullerton). Vertical scale is arbitrary. Curves were separated for clarity. Curves 1 and 2, 3 and 4, and 5 and 6 retain their original count-rate relations.

sufficiently distinctive to give an apparently unique gamma-ray spectrum of most samples. The possibility of coincidental spectra from soils of different origin, however, is appreciable. The Linker and Fullerton soils differ in texture and color and were collected from sites at least 50 miles apart. The Linker soil is formed from interstratified sandstone and shale of the Pottsville formation of Jefferson County, Ala. The Fullerton is a cherty silt loam weathered from the Chepultapec limestone and Copper Ridge dolomite of the same county. The spectra, however, are nearly identical. This finding introduces some doubt as to whether gamma-

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ray spectra of irradiated soils may be used reliably as an aid to soil classification or as a means to trace soil specimens to the location where they were collected when the conditions of measurement are as specified.

Gamma-ray spectra of several soils in this study are shown in Fig. 3, after 62 days' decay. These spectra are

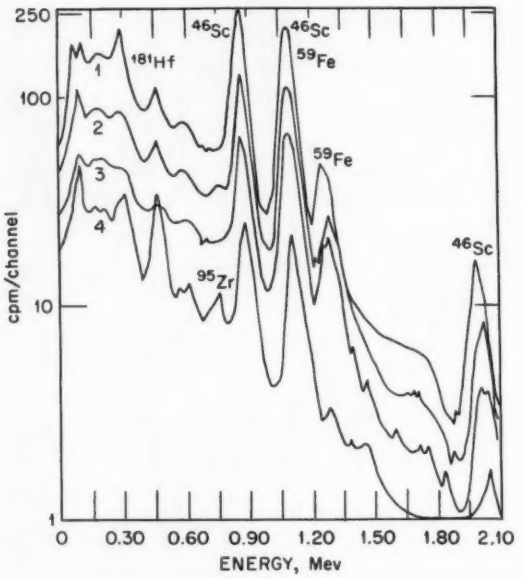

Fig. 3 Gamma-ray spectra of soils taken 62 days after 5-min irradiation with thermal neutrons at a flux of 2×10^{13} n/(cm^2)(sec). Curve 1, Singapore; 2, Minnesota (Fayette); 3, Lebanon; 4, New Zealand.

dominated by ^{46}Sc and ^{59}Fe, and some of them have remarkably similar structures, to have arisen from samples of such diverse origin. In some soils certain nuclides (e.g., ^{181}Hf) appear and give the corresponding spectrum a distinctive appearance. In many of the soils, however, scandium and iron occur in sufficiently constant ratios to give the spectra a very similar appearance. It is not implied that the spectra of longer-lived nuclides of all soils are similar; rather, it is pointed out that, if even a few are similar when obtained from material of dissimilar origin, then there is a possibility of error in matching specimens or tracing them to their origin.

Quantitative Analysis of Soils

Some of the nuclides shown in the spectra were determined quantitatively for all soils (Table 3). These

Table 3 Element Content of Soils as Determined by Nondestructive Neutron Activation Analysis

Soil	Fe, %	Sc, ppm	La, ppm	Co, ppm	Eu, ppm	Sm, ppm
Lebanon	1.4	3.6	6.4	4.7	0.3	1.7
Malaysia	2.7	9.1	*	1.0	*	0.6
New Zealand	0.2	1.3	*	*	*	0.6
Hawaii	19.2	25.1	17.5	38.6	1.3	5.2
Norway	2.0	2.6	2.1	2.3	0.1	†
Tokyo	6.4	24.4	5.7	27.3	1.1	†
Brazil	0.9	1.8	4.1	0.4	0.1	†
Australia	5.9	8.8	22.7	7.3	1.0	†
Southern Rhodesia	9.6	41.0	9.7	64.9	1.5	†
Linker	3.0	3.4	20.7	3.7	0.7	†
Fullerton	2.2	2.6	17.3	1.9	0.5	†
Fayette	1.9	6.8	18.0	8.1	0.7	4.3
Kilkeny	3.4	8.0	23.6	9.0	0.8	4.5

* = not detected, † = not determined.

results are presented to illustrate the potentialities of nondestructive neutron activation analysis on a highly diversified group of soil samples. They were obtained by the solution of simultaneous equations (^{59}Fe, ^{46}Sc), sum-peak analysis (2.5-Mev sum peak of ^{60}Co), and peak-height analysis (^{140}La, ^{153}Sm, ^{152}Eu) according to the method of Covel.[8] Values for iron, scandium, and lanthanum are most precisely determined and have one standard-deviation counting error of less than 10%. Cobalt is the least precisely determined and has errors ranging from 100% for the smallest amount shown to 5% for the largest. Europium and samarium values are based on total counts, ranging from 320 to over 10,000, and have errors ranging downward from about 15% for one standard deviation.

Nuclides not measured quantitatively in the spectra include ^{24}Na, ^{181}Hf, ^{95}Zr, ^{82}Br, and ^{76}As. Some were not measured because they have nearly overlapping photopeaks in the region of 0.55 Mev, which cannot be resolved by the detector system used; ^{82}Br has other photopeaks, but these are obscured by photopeaks of other nuclides; ^{181}Hf and ^{95}Zr were not anticipated and were not found until the data were plotted at the end of the experiment, although they could be measured by Covel's method (Ref. 8); ^{24}Na could easily be measured, but in these experiments there seemed little to gain by doing so since it was present in large quantities and presented little opportunity to discriminate among samples.

Much of the apparent similarity among the gamma-ray spectra is the result of the constant relation between scandium and iron (Fig. 4*a*).

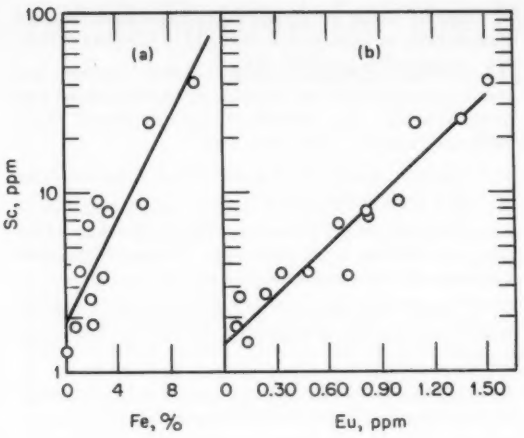

Fig. 4 Correlations between scandium and other elements in a worldwide collection of soils. Curve *a*, Sc–Fe, $r^2 = 0.80$. Curve *b*, Sc–Eu, $r^2 = 0.89$.

The relation is logarithmic, with an r^2 value of 0.8, which indicates that the geochemical behavior of these elements might be quite similar under a variety of weathering conditions. The relationship between scandium and cobalt, another member of the first transition series of metals, is shown in Fig. 5. The correlation is linear, with an r^2 value of 0.96.

Scandium is sometimes included in discussions of the rare-earth elements because it occurs in the same periodic group, although Haskin and Frey[9] indicate that its behavior is considerably different from the rare earths. Correlations between scandium and europium (Fig. 4*b*) were, nevertheless, highly significant with an r^2 value of 0.89. The data on samarium are too scanty to attempt similar correlations. However, the data in Table 2 indicate a probable relationship between samarium and europium. It was found that lanthanum, the first member of the rare-earth series, was not strongly related to the other members of either the rare-earth series or the first transition series of metals in these samples.

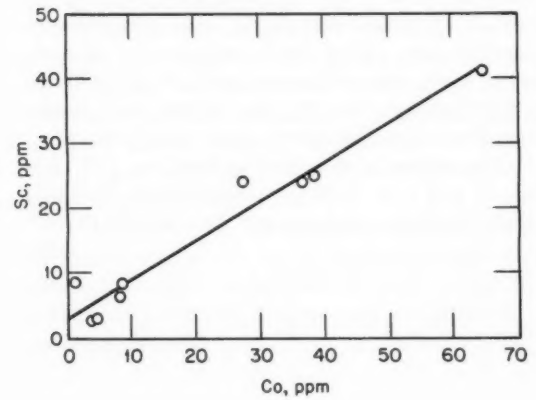

Fig. 5 Correlation between scandium and cobalt in worldwide collection of soils. Five points with scandium and cobalt values each below 5 ppm are omitted for clarity. Correlation coefficient $r^2 = 0.96$ was obtained for 14 observations.

CONCLUSIONS

Many of the elements of soils which can be neutron activated and which have appreciable half-lives are members of either the first transition series of metals or the rare-earth series. These elements have somewhat similar geochemical behavior, and this can result in quite similar gamma-ray spectra when soils are analyzed by neutron activation.

When attempts are made to match specimens of material by neutron activation methods, it is assumed that the trace-element components of the material are more or less randomly distributed and that this distribution is not likely to be duplicated in materials of diverse origin. The relationships that have been shown make this assumption of doubtful validity for soils analysis. This doubt is reinforced by the finding of two soils in Alabama which gave practically identical gamma-ray spectra. It is concluded that gamma-ray spectra of soil subjected to high-flux irradiations and measured with a NaI(Tl) crystal and 400-channel analyzer should not be used without support from other evidence for matching specimens in criminal investigations or soil correlation studies. It is recognized, however, that low-flux or cadmium-shielded irradiations could broaden the usefulness of nondestructive activation analysis, particularly if the spectra are obtained with the recently developed high-resolution crystals.

This method of analysis has permitted the determination, for example, of Sc, La, Eu, and Sm—elements not commonly investigated in soil studies. Convenient alternative methods to determine these elements are not available, although they can be measured with low sensitivity by spectrographic methods. Instrumental activation analysis can be recommended to measure these elements in soils when NaI-crystal detectors and 400-channel analyzers are used. Broader applications depend on the availability of high-resolution crystals and high-capacity multichannel analyzers for gamma-ray spectrometry. (PSB)

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Radiotracer Method for Measuring the Abrasivity of Magnetic Recording Tapes

By John D. Buchanan*

A radiotracer method for measuring the abrasivity of magnetic recording tapes was developed by Isotopes and is being used in government and industrial laboratories. A recording head made of Kovar containing cobalt is used. This head is reactor-irradiated to produce ^{60}Co, and the amount of this radioisotope transferred to the tape indicates the amount of Kovar worn from the head and thus the abrasivity of the tape itself.

In the development work, 200-ft-long samples of 0.5-in.-wide instrumentation tape were tested using the same tape wrap angles, tensions, and speeds as those used in actual recorders. The precision of 5-pass tests was about 4%, with virtually all the abraded radioactive material adhering to the tape. Abrasivity was found to be directly proportional to tape tension but independent of tape speed. Relatively large differences in abrasivity were found, even among samples from the same reel of tape. Comparison of measurements made with radioactive heads of two different designs indicates that the test method gives relative abrasivity values applicable to actual recorder heads.

The method is described in detail by J. D. Buchanan and J. D. Tuttle, in *International Journal of Applied Radiation and Isotopes*, **19**: 101-121 (1968). (MG)

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A Method for the Nondestructive Detection of Moisture in Roof Sandwiches

By Ralph Lahmon*

Abstract: *A nondestructive method for measuring the moisture content of sandwiched roofing material has been developed which enables a rapid inspection to be made without puncturing the roof. An instrument normally used to measure moisture content in soils was standardized for this application. Field testing showed the instrument to be capable of indicating areas of moisture invasion. The gage has been successfully used on several building roofs to locate and map areas that were seriously saturated with moisture. In a few cases it was used to relate the deterioration of the roof to age and to indicate where either replacement or resurfacing of certain roof areas was required. The gage will be used for troubleshooting, and savings of several thousand dollars a year are expected. A program is being developed in which the gage will be used routinely to inspect the roofing sandwiches.*

In the past few years about 45 thousand sq ft/year of water-damaged roofing sandwiches have been replaced in the Eastman Kodak plant at Rochester, N. Y., and conceivably this rate could increase to 100,000 sq ft/year in 5 years with increased installed roof area. Occasionally, expense above that of roof replacement was incurred when the moisture caused damage to the interior of the building or repairs interrupted production. Since this water invasion generally starts at a fairly localized source, an inspection system that would reveal the early accumulation of water and permit accurate mapping of the affected area could considerably reduce repair costs.

Previously used methods for inspecting a roofing sandwich required removal of a section of the sandwich for inspection and subsequent determination of the moisture content in a laboratory. This procedure is expensive and not practical for any extensive inspection program since about 1 hr of labor is required to cut and patch each sample location.

The purpose of the presented project was to determine whether or not a density-moisture gage designed for field testing of soils could be used for rapid, nondestructive measurement of moisture in roofing sandwiches.

DESCRIPTION OF GAGE

The instrument tested is a combination density-moisture gage with two measurement channels—i.e., a moisture channel and a density channel—that operate independently of each other. A 4.5-mc Ra–Be source is used and is common to both channels since it emits both neutrons and gamma radiation. The neutrons are used for the moisture determination and the gamma radiation for the density measurement. The moisture-measurement channel incorporates two boron trifluoride tubes that detect neutrons but are insensitive to gamma radiation. The density channel uses two halogen-quenched detectors that detect gamma radiation and are completely insensitive to neutron radiation.

The Ra-Be source emits "fast," i.e., high-energy, neutrons. As the emitted neutrons penetrate the test material, they collide with the atomic nuclei of the elements of the test material and scatter in all directions. When a neutron collides with a nucleus, the neutron loses some of its energy, the amount depending upon the particular nucleus with which it collides. The amount of energy lost is a function of the ratio of the mass of the neutron to the mass of the nuclide with which it collides and of the angle of reflection. For example, when a neutron collides with a hydrogen nucleus, which has approximately the same mass as a neutron, the neutron loses more of its energy than when it collides with the nuclei of heavier elements. Thus the higher the hydrogen content in the test material (water, for example, is two-thirds hydrogen atoms), the higher the number of "slow" neutrons that will be scattered back to the detectors. For each slow neutron that enters the detector, an electrical impulse is produced and transmitted to a counter. In a homogeneous material the counting rate is directly proportional to the hydrogen content over a range of approximately 5 to 30 lb of moisture per cubic foot.

The density measurement depends on the backscattering of the gamma radiation by the test material; hence the detectors are shielded so that only the gamma radiation scattered from the test material reaches the detectors. The gamma radiation entering the material is scattered in all directions. This scattering continues until the gamma ray is completely absorbed or emerges from the material. As the mass per

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unit volume of the test material increases, the scattering power of the medium increases proportionately. With each scattering process, however, the gamma ray loses energy. Thus, although an increase of density in the material increases the probability of scattering of the gamma ray, the probability that the gamma ray will be absorbed before it reaches the detector is also increased. The combined effect of these probabilities is that fewer gamma rays per unit time reach the detectors as the density of the test material increases. This inverse relationship between counting rate and density is linear over a range of approximately 60 to 160 lb/cu ft.

Handling and Operation of the Gage

Since the density–moisture gage contains radioactive material, a safe handling and operating procedure was developed to ensure that neither the operator nor stored photographic products were needlessly exposed to the radiation. As part of the preparation of the safe operating procedures, tests were made to determine the possible effects on sensitized products of the radiation from the gage while being used for roofing inspections. The most important data obtained from these tests were the maximum time periods that the gage could be used at a specified distance from sensitized products when shielded by various thicknesses of concrete roof slab. These data were the basis for the safe handling and operating procedures.

METHOD OF INVESTIGATION

A set of reference readings was taken in the laboratory with the density–moisture gage of samples of the various types of roofing sandwiches used in Kodak Park. Separate pieces of the various components of a roofing sandwich (insulation, vapor barrier, and roof membrane) were assembled to duplicate the various roofing sandwiches. Then gage readings were taken on these laboratory samples (representing new, dry roofs), and the readings from the individual components were tabulated for future reference.

The effect of the presence and the location of moisture in the roofing sandwich on the gage reading was determined by mock tests made in the laboratory using wet insulation. Two 1-in.-thick pieces of fiberboard insulation were soaked in water and weighed before and after soaking to determine the amount of moisture absorbed. Then gage readings were taken on 2 in. of wet insulation and combinations of 1-in. dry and 1-in. wet insulation. In one case the wet piece was on the top and the dry piece on the bottom; then the reverse case was tested with a wet piece on the bottom and a dry piece on the top.

As a final evaluation the gage was field tested on several buildings where the roofing sandwiches were known to have wet areas. The gage was used to map wet areas; then two inspection samples were cut from the roof to verify the gage readings. One cut was made at the highest gage reading and one in the middle of the counting range. These sample sections were inspected to determine what deterioration of the material had occurred and to locate the position of the moisture in the roofing sandwich.

RESULTS

Table 1 lists the various roofing sandwiches that were tested in the laboratory and the corresponding readings to be used as a "standard" for these sandwiches. The sandwich structures included different types of insulation with and without a vapor barrier (2-ply asphalt-saturated felt) and with and without a gravel surface. In all tests, four plies of 15-lb asphalt-saturated felt were used for the roof membrane. A few

Table 1 Standard Reading for the Various Roofing Sandwiches

Deck*	Vapor barrier (felt)	Roof membrane (15-lb felt)	Gravel surface	Gage reading, cpm (±5%)
4-in. concrete	2 ply	4 ply	No	660
4-in. concrete	2 ply	4 ply	Yes	530
4-in. concrete	None	4 ply	No	550
4-in. concrete	None	4 ply	Yes	430
4-in. concrete	2 ply	4 ply	No	620
4-in. concrete	2 ply	4 ply	Yes	500
4-in. concrete	None	4 ply	No	510
4-in. concrete	None	4 ply	Yes	400
4-in. concrete	2 ply	4 ply	No	510
4-in. concrete	2 ply	4 ply	Yes	380
4-in. concrete	None	4 ply	No	530
4-in. concrete	None	4 ply	Yes	420
4-in. concrete	2 ply	4 ply	No	280
4-in. concrete	2 ply	4 ply	Yes	200
4-in. concrete	None	4 ply	No	250
4-in. concrete	None	4 ply	Yes	180
Metal	None	4 ply	Yes	170
Metal	None	4 ply	Yes	210
Metal	None	4 ply	Yes	210

*A 4-in. granite block was used to simulate the 4-in. concrete deck.

tests were run with a metal deck, but, in most cases, a 4-in. granite block was used to simulate a 4-in. concrete deck.

Readings taken for the individual materials or components of a roofing sandwich (Table 2) indicate the potential effect that each material will have on the gage readings when it is used in a roofing sandwich. For example, the granite block alone had a reading of 160 counts per minute (cpm) whereas 2 in. of fiberboard insulation plus the granite block had a rate of 170 cpm. This is compared to a count rate of 500 cpm for a 4-ply bitumen-bonded roofing membrane on the granite block. A gage reading of 510 cpm was obtained when these same components were assembled to form a simple roofing sandwich.

Table 2 Readings of Different Material Used in Roofing Sandwiches

Material	Reading, cpm (±5%)
4-in. granite block	160
4-in. granite block + 1-in. fiberboard	160
4-in. granite block + 2-in. fiberboard	170
4-in. granite block + 2-in. foam glass	100
4-in. granite block + 2-ply felt (vapor barrier)	230
4-in. granite block + 4-ply felt (roof membrane)	500
4-in. granite block + 6-ply felt	920
4-in. granite block + 2 ply + 1-in. fiberboard	260
4-in. granite block + 2 ply + 2-in. fiberboard	270
4-in. granite block + 2-in. fiberboard + 2 ply	270
4-in. granite block + 2-in. fiberboard + 4 ply	510
4-in. granite block + 2 ply + 2-in. fiberboard + 2 ply	400
4-in. granite block + 2 ply + 2-in. fiberboard + 4 ply	620
4-in. concrete block (cured in open shed 2 weeks)	270

Table 3 shows the results of laboratory tests made on roofing sandwiches with wet insulation. The insulation was a 1-in.-thick fiberboard which contained 2 lb of water per pound of fiberboard.

Table 4 shows the readings that were obtained in field testing the gage on several buildings. These readings were verified by making two inspection cuts per building.

DISCUSSION

The gage readings for the roofing sandwiches tested in the laboratory (Table 1) varied from about 200 to 700 cpm. A large percentage of this "dry" count can be attributed to the bitumen used in applying the felts. The bitumen is a hydrocarbon material with a high percentage of hydrogen atoms. The readings taken on the individual components of a sandwich illustrate this effect (Table 2). Also, attenuation by the gravel surface used on the laboratory-tested samples reduced the readings by about 20%. This means that the "standard" readings obtained from the laboratory test are not unique values and that actual roofing sandwich installations will vary depending on the thickness (amount) of bitumen used and the thickness of the gravel surface.

The laboratory tests show that the gage's sensitivity to moisture decreases with distance of the moist spot location from the gage. This is illustrated by the readings (Table 3) that were taken using a wet piece

Table 3 Readings for Wet Insulation (2 lb of Water per Pound of Material)

Deck (4-in. granite block used)	Vapor barrier (felt)	Insulation (fiberboard)	Roof membrane (15-lb felt)	Gravel	Reading, cpm (±5%)
4-in. concrete	2 ply	2 in. wet	4 ply	Yes	2790
4-in. concrete	2 ply	2 in. wet	4 ply	No	3400
4-in. concrete	2 ply	1 in. dry and 1 in. wet (top)	4 ply	No	2800
4-in. concrete	2 ply	1 in. wet and 1 in. dry (top)	4 ply	No	1830
4-in. concrete	2 ply	2 in. dry	4 ply	No	620
4-in. concrete	None	2 in. wet	4 ply	No	3440

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Table 4 Results of Field Test on Several Buildings

Deck	Vapor barrier	Insulation	Roof membrane	Surface	Age, years	Readings, cpm
Concrete	1-ply felt	2-in. fiberboard	4 ply	Slag	40	220 → 2300
Concrete	Koroseal	2-in. Fiberglas	4 ply	Smooth	10	300 → 2000
Concrete	None	Cinders	4 ply	Smooth	45	540 → 1500
Concrete	None	1-in. cork	4 ply	Smooth	30	200 → 2800
Concrete	None	2-in. cork	4 ply	Gravel	25	500 → 2500
Concrete	1-ply felt	2-in. Celotex	4 ply	Slag	25	750 → 3800
Metal	None	1½-in. fiberboard	4 ply	Gravel	New	120 → 370

and a dry piece of insulation in a roofing sandwich. With a wet piece for the top inch and a dry piece for the bottom inch of insulation, the gage reading was 2800 cpm. When the pieces of insulation were interchanged, the gage reading was 1830 cpm. This means that the location of moisture in a roofing sandwich has to be considered when the operator interprets the gage readings to determine the degree of moisture saturation. The pieces of insulation used in these tests were fiberboard, and the wet piece contained 3 lb of water per 120 cu in.

The field tests showed that the variation in the location of moisture did not create a serious problem in evaluating existing roofing sandwiches. First, if the insulation medium is continuous in depth, the moisture will be fairly uniformly distributed throughout the insulation. In the cases where the insulation consists of two pieces separated by a bitumen bonding layer, only one layer—usually the top piece—will contain moisture. Second, the measurement of moisture in a roofing sandwich does not need to be exact to evaluate the condition of the sandwich. That is, for a particular roofing sandwich that has a standard counting rate of 500, readings around 800 would indicate that moisture was present, but not enough to justify replacing the existing roofing sandwich. On the other hand, a gage reading of 1500 or higher would indicate that the insulation has become saturated with moisture and is very likely to have deteriorated to the point where replacement is necessary. When readings are in the 700-to-1300 range, engineering judgment has to be used to determine whether the economics justify replacing the entire sandwich or only resurfacing the roof membrane to prevent further moisture accumulation.

One interesting discovery from the use of the gage in these field tests is that the source of the moisture can also be determined. In mapping out a wet area of a roofing sandwich, the location of the highest readings will be found at the origin of the moisture migration. In the building roofs that were field tested, the source of moisture was usually at the brick penthouses or concrete piers, probably because moisture travels through the porous brick or concrete material to the roofing insulation.

Disposition and Application

The density-moisture gage has been used quite successfully in locating and mapping wet areas on several roofs. To further increase the speed of the inspection procedure, a different readout instrument was purchased. This instrument gives immediate and continuous readings in cpm. This means that the gage can be mounted on a cart with only a fraction of an inch clearance between the gage and roof membrane and can be moved freely across the roof. The mobility of the gage will enable a faster inspection than the 3-min test procedure required for the digital readout originally purchased with the gage. Some accuracy of the readings will be sacrificed by using the rate meter but not enough to affect seriously the interpretation of the readings (see Appendix).

The gage will be used for "trouble shooting" inspection of roofing sandwiches for suspected damage areas; meanwhile a proposal for inspecting roofs routinely is being developed. Under this program the gage would be used in the routine inspection of all components of all roofs. To what extent the gage will be used in this program depends on the success of the plans to make the gage mobile.

APPENDIX

Figure 1 shows the cart that was built for transporting the density-moisture gage. The tray on which the gage is positioned (Fig. 1) is adjustable for minimum clearance between the gage and the roofing membrane.

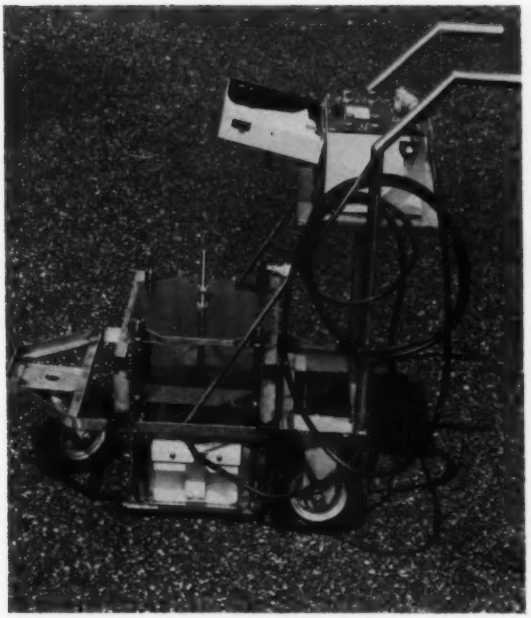

Fig. 1 Cart for transporting the density–moisture gage.

The gage mounted on the cart was used in an inspection of a processing laboratory roof. The area of the roof is about 50,000 sq ft. Of this area approximately 5000 sq ft (10%) was directly inspected with the gage in 2 days. This is a higher percentage of a roof area than would be inspected on a building in Kodak Park because the building was in a remote location.

Figure 2 shows the ponded water that had accumulated on the processing laboratory roof. The presence of this "surface" water did not affect the use of the gage to inspect the roofing sandwich for moisture. By keeping the gage essentially on the roofing membrane, the water is completely displaced by the gage. The gage is sensitive to the medium directly beneath and is basically insensitive to the surrounding medium.

Figure 3 illustrates the technique used in mapping a wet area and shows one of the wet areas that was located and mapped. In this case a typical dry reading is about 300 cpm. To verify the presence of moisture in the sandwich, a sample cut was made (note black patch) at a point where the reading was 2500 cpm. The insulation that was removed from the cut was completely saturated.

Fig. 2 Accumulation of ponded water on laboratory roof.

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Fig. 3 Location and mapping of wet roof areas.

Discharge Measurements Using Radioisotopes in High-Head Turbines and Pumps

By R. L. Hansen*

Abstract: *A method, using ^{82}Br, being developed for measuring flow rates in high-head turbines and pumps was tested on a 6000-ft-long, 6-ft-diameter penstock and a 6000-ft-long, 8-ft-diameter pump discharge line. The data obtained were less precise than desired but were encouraging and indicated pipe lengths required for natural turbulence to mix pipe flow and tracer and for additional mixing induced by a pump.*

A research program jointly funded by the Bureau of Reclamation and the Division of Isotopes Development of the U. S. Atomic Energy Commission is directed toward perfecting a precise and relatively simple method of measuring flow rates in large high-head equipment, such as turbines and pumps, by using radioisotopes.[1] Present standard techniques for making such measurements do not yield the desired accuracy, especially for acceptance testing. However, knowledge of methods and instrumentation developed in previous radioisotope studies[2] for measurement of open-channel flow rates is adaptable in part to this program. Research conducted at the Flatiron Power and Pumping Plant (Loveland, Colo.) on methods to improve the reliability and accuracy of the radioisotopic measurement systems is reported here.

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TEST PROCEDURES

The radioisotope tracer method was tested by measuring the discharges from a 6000-ft-long (1830 m), 6-ft-diameter (1.8 m) penstock that feeds the Flatiron turbine and from a 6000-ft-long, 8-ft-diameter (2.6 m) pump line from a pump-turbine unit. The radioactive tracer in samples taken from the flow was determined by the total-count technique.[3]

The flow measurements made with radioisotopes were compared with those made by calibrated spiral-case flowmeter taps at the turbine. Repeatability of the measurements was of first concern, and the spiral-case flowmeter gave an independent measure of discharge and indication of the steadiness of the flow.

Specialized equipment for these tests included injection and sampling probes that could be located at various positions in the penstock diameter. A mechanical system provided for either injection of the radioisotope or sampling of the tracer–water mixture at points between the wall and the center line of the penstock. The position of the injector and sampler in the pipe was established using a threaded bronze stem passing through a handwheel and bronze nut, all supported by four steel legs attached to the manway cover.

Turbine Tests

In the turbine-discharge measurements, radioisotope tracer was injected essentially at the pipe wall (0.04 radius), at 0.1 and 0.4 of the pipe radius from the pipe wall, and at the center line, these positions being selected to show the effect of the radial location of single-point injection upon mixing length. Thirty-five separate injections of $K^{82}Br$ in water solution were made, resulting in 82 computations of the discharge. Samples were taken in the penstock at lengths of about 47, 311, and 645 pipe diameters downstream from the injection as well as at the turbine draft tube. These lengths were chosen to show the results of mixing the tracer and flow for lengths well above and well below the computed minimum mixing length of about 100 pipe diameters. The measurements at the inlet and outlet points in the draft tube were to explore any unique effects of the turbine on mixing or other aspects of the measurement.

Simplified pressure reducers, developed in hydraulic studies,[3] were used. Samples were taken at pressures of 350 psi (24.5 kg/cm^2) and 475 psi (33.3 kg/cm^2) at a rate of 20 to 25 gal/min and passed through a 1.8-cu ft tank (Fig. 1) containing a scintillation

Fig. 1 Sample tank containing detector and connected to penstock sampling system. Small plastic tube at entrance to tank used to collect samples for "integrated sample" and "dilution" methods of discharge measurement.

counter to measure the radioactivity of the tracer–water mixture. Tracer was also injected into the pipe through four configurations of holes in the injector tips at pressures ranging from 295 psi (20.7 kg/cm^2) to 950 psi (66.5 kg/cm^2) in order to explore the possibility of using high-pressure injection to decrease the mixing length.

The radioisotope solution was forced into the penstock by a diaphragm pump or a gas-pressurized pulse system. The pump operated at 230 strokes per minute from a 110-volt 60-Hz portable generator. The pump capacity was 9000 ml/hr at 0 psi (maximum) and 5600 ml/hr at 5000 psi. Flow rates through the pump were varied by changing the length of stroke of the piston driving the diaphragm. The pulse-injection system was constructed of 1800-psi sample cylinders and 3000-psi piping and valves (Figs. 2 and 3). A 3.78-liter cylinder held water to purge the tracer cylinder after injection, and the piping was arranged to apply regulated gas pressure from a 2000-psi nitrogen or compressed-air cylinder to the 1-liter tracer cylinder and then to the water purge for rinsing. A combination of flexible and rigid stainless-steel tubing carried the tracer solution and then the water to the injector. The rate of tracer injection could be changed by increasing the differential pressure between the nitrogen supply and the penstock.

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Pump-Turbine Tests

In a second series of measurements, the pump-turbine unit was operated as a pump to evaluate procedures and techniques used in the radioisotope method applied to pump flows. The injection and counting equipment was similar to that used in the turbine-feed tests. Injections of $K^{82}Br$ were made in the pump flow at a distance of 6 in. from the wall of the inlet elbow upstream from the pump and 8 in. from the pipe wall downstream from the pump and a butterfly valve. Mixing lengths of 164 and 755 diameters were tested using a system containing two tubes for sample withdrawal and a counting unit located 1300 ft downstream from the pump and a second sampling and counting system for taking a sample through the sidewall of a valve located 6000 ft downstream. A total of 27 discharge computations were made from the 12 injections.

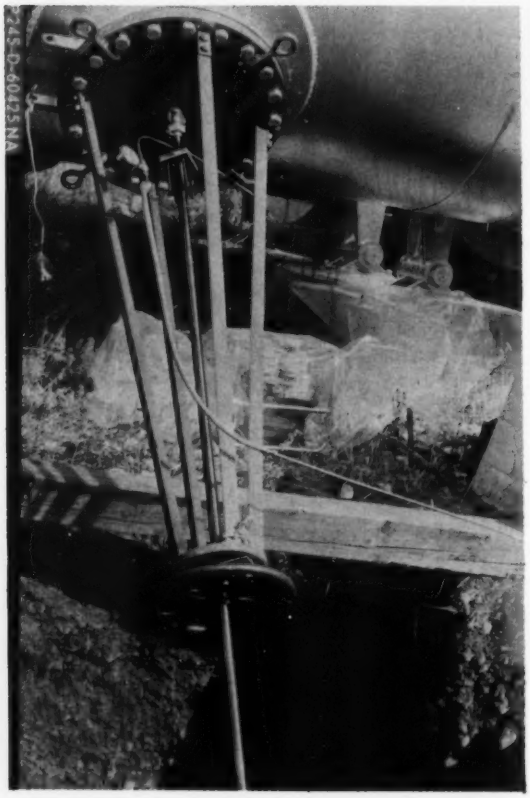

Fig. 2 Injection tube inserted in penstock and connected to radioisotope supply system. Manway cover located 135° from vertical when observed looking downstream.

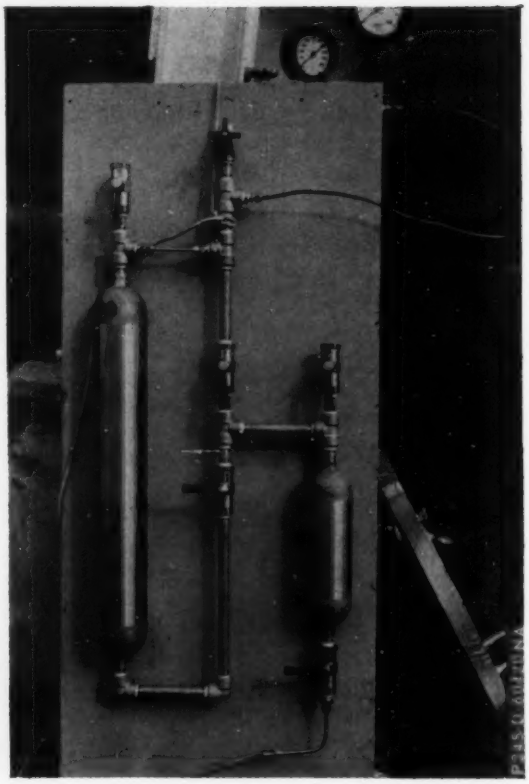

Fig. 3 Radioisotope supply system for pulse injection using compressed nitrogen or air.

RESULTS

In general, the discharge measurements (Table 1) on neither the penstock nor the pump were as precise as desired, but they gave good information on the lengths of pipe required for natural turbulence to mix the pipe flow and tracer injected at the pipe center line. The data also indicated the suitability of both the radiation-measurement and the injection and sampling techniques. Injections at a selected point between the center and side of the penstock did not produce a significant decrease in the mixing length.

Mixing of the tracer and pipe flow was increased by the pump. A mixing length of 164 diameters in the 8-ft-diameter pump line appeared to be satisfactory for a tracer injection near the pipe sidewall into water flowing at about 6 ft/sec (1.8 m/sec). In a 311-diameter mixing length with a centerline injection into water flowing about 4.5 ft/sec (1.4 m/sec) in the

Table 1 Summary of Discharge Measurements

Measurement length, diameters	Sample	No. of measurements	Average discharge, cu ft/sec		Deviation,* %
			Flowmeter	Radioisotope	
		Penstock			
46.5	Center	12	127.0	131.5	+3.5
	Side	12	127.0	94.8	−25.4
310.8	Center	23	126.7	130.7	+3.2
	Side	23	126.7	131.3	+3.7
645	Side	12	127.2	128.7	+1.2
		Pump			
164	0.07-radius	11	287.6	281.3	−2.2†
	0.57-radius	11	287.6	282.3	−1.8†
755	Side	5	288.2	289.9	+0.6

*From flowmeter results.

†The first 6 of the 11 measurements averaged about 5% lower than the flowmeter. They are believed to be in error and lower the average values. The 5 remaining measurements gave values about 2.1% higher than the flowmeter.

turbine penstock, the natural turbulence did not produce satisfactory mixing.

Some of the discharges computed from the samples taken at the draft tube of the turbine were obviously in error, and the measurements had a greater variation than desirable. Investigations using Pontacyl Pink Dye and a fluorometer showed that a significant amount of recirculation of water from the tailrace into the draft tube was occurring for the less-than-maximum discharge used in the measurements. The recirculation caused excessive dilution of the sample being extracted from the draft tube and thus an error in the discharge measurements. It is therefore recommended that sampling from turbine draft tubes be avoided whenever possible unless it can be shown that no recirculation up the draft tube is occurring.

When the configuration of the holes in the injection tip was varied, injection pressure did not show a significant effect on the mixing. Discharges computed for a mixing length of 46.5 diameters, selected to more readily distinguish the change in mixing, differed from flowmeter values by +10 to −30%. (MG)

REFERENCES

References marked with an asterisk have not been verified by the editors; for further explanation, see the Foreword.

1. J. C. Schuster and R. L. Hansen, Discharge Measurements Using Radioisotopes in High-Head Turbines and Pumps at Flatiron Power and Pumping Plant, Colorado Big Thompson Project, Bureau of Reclamation, General Report No. 40, December 1968.
2.* R. L. Hansen, Radioisotope Flow Measurements on the Friant-Kern Canal, Central Valley Project, Fresno, California, Bureau of Reclamation, Division of Research, Report No. ChE-41.
3. B. A. Fries, Gas-Flow Measurements by the Total-Count Method, *Int. J. Appl. Radiat. Isotop.*, 13: 277-280 (1962).
4.* J. W. Ball and W. P. Simmons, Progress Report on Hydraulic Characteristics of Pipeline Orifices and Sudden Enlargements Used for Energy Dissipation, Bureau of Reclamation, Division of Research, Report No. Hyd-519.

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Some Potential Uses of Radioisotopes for Security and Law Enforcement*

By James Wing†

Abstract: *The results of feasibility studies on several applications of radioisotopes for security and law enforcement are reported. Techniques and systems have been developed for secret marking of equipment with radioactive screws, invisible labeling and storing of secret information by recoiled radioisotopes, treating hydrogenous surfaces with tritium for positive identification, safe-locking by radioisotope keys and gamma-ray spectrometry, and detecting door intrusion by gamma-ray spectrometric monitors. The advantages and limitations of each potential use are discussed.*

Nuclear radiation below extremely high levels cannot be detected directly by any of the five senses. Hence it is particularly suitable for any application that requires some element of secrecy. The results of a series of feasibility studies on the use of radioactivity for security and law enforcement, including the use of radioisotopes for invisible marking and of nuclear radiations as secret transmitting signals, are reported. The presently developed techniques and systems are not necessarily superior to the many others now in existence. The advantages, limitations, and precautions in these applications are discussed.

SECRET LABELS BY RECOILED RADIOISOTOPES

Secret labeling[1] of solid surfaces was made with radioactive fission fragments that were emitted from a spontaneously fissioning material. The fission fragments were collected on the surface to be labeled through a steel stencil placed between the source and the surface. The source used was 1.5 μg of ^{252}Cf, which was deposited in a round hollow that was 5 mm in diameter and 1.5 mm deep and covered with 200 μg of aluminum per square centimeter. The distance between the bottom of the hollow and the surface to be labeled was about 3 mm. The source was shielded with 10 cm of boron-doped paraffin to stop neutron emission.

The resulting label measured 2 by 3 mm and was not directly observable but could be revealed by autoradiography. The amount of the recoiled materials was so small that most other presently known methods of detection would not be able to detect the presence of the label. The recoiled fission fragments carried a considerable amount of kinetic energy. Hence the label was firmly embedded and was not removed by repeated washings with water, detergent solutions, alcohol, or acetone, or even by flame (i.e., labels on inflammable surfaces), short of destroying the labeled surface itself.

Figure 1 is a log-log plot of the gross beta activities of the collected fission fragments, measured with an end-window gas-flow proportional counter, vs. days after the end of collection. A calculated curve by Bolles and Ballou[2] is also included for comparison.

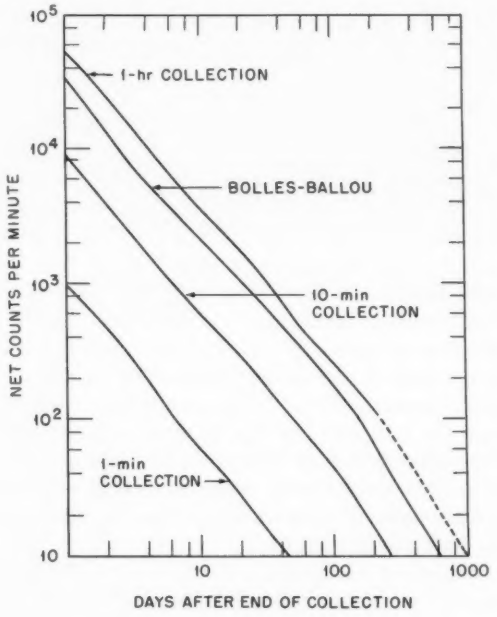

Fig. 1 Beta activities of fission fragments as a function of time after the end of collection. The curve labeled Bolles–Ballou is a calculated one (Ref. 2).

Autoradiography of the labels was made with Ilford Industrial G X-ray films. Exposure times varied

*Based on work supported by the U. S. Atomic Energy Commission.

†National Bureau of Standards, Washington, D. C. 20234

from 0.5 to 24 hr, depending upon the amount of beta activity present in the label. Since the fission product was a mixture of isotopes with different half-lives, labels could be made to last for almost any desired period of time. The lifetimes of the labels were determined by repeatedly autoradiographing the labels with exposures of 24 hr each. The labels formed with 1, 5, and 10 min of collection were detectable up to 14, 60, and 110 days, respectively, after the end of collection. The limit of detection was 30 cpm above background. The lifetime of a label made with a 1-hr collection at this limit was estimated from the extrapolated decay curve (dashed line) in Fig. 1 to be 1 year.

Eighteen different metals were exposed to the ^{252}Cf neutrons for 1 hr each, but no induced radioactivities were detected. No radiation damage to the labeled objects was apparent or expected. One hour after the end of an hour's collection, the beta activities on the label constituted only 1.5% of the maximum permissible level—i.e., 10 μc of fission products in total body.[3]

The present technique of secret labeling has an advantage over the use of self-evaporating radioactive inks in that the fission-fragment labels are not removed by washing with noncorrosive solvents. No visible impressions are ever made on the labeled surfaces, since there is no physical contact of the source of the labeling materials with the surfaces to be marked. The label may be applied to any nonsticky surfaces having enough flat and smooth areas free of loose materials such as dust. Eventually—depending upon the amount of the fission fragments collected—the radioactivities of the label will decay to a level undetectable by any means. At present, ^{252}Cf is available at $100 for 0.1 μg.

STORAGE OF SECRET INFORMATION

As an extension of the above technique, a method was developed for temporary storage of secret messages. A version of the technique using the recoiled fission fragments from ^{252}Cf has already been reported.[1,4] Besides ^{252}Cf, about 50 μg of ^{232}U, in equilibrium with its decay products, has also been used as the source of recoiled materials.

An invisible message was formed by catching the recoiled radioisotopes, through different stencils, onto a catcher foil. The message was revealed by the autoradiographic process as described above (Fig. 2). One week after the collection, the message was

Fig. 2 Autoradiograph of a message (made with a 3-min collection from ^{232}U for each letter) 48 hr after the collection and after a 24-hr exposure with the Ilford film.

undetectable; thus there is a built-in self-destruction feature.

For lengthy messages, the efficiency of recoil collection can be increased by using a source of higher strength and by collecting in vacuum or helium jets and on negatively charged metallic foils. Simultaneous storing of different parts of the entire message with several ^{232}U sources and a steady-state operation of sending out short portions of the message, one at a time, are feasible.

Because the beta decay of fission fragments from ^{252}Cf follows an almost linear log-log relationship (Fig. 1)—that is, they are long-lived—^{252}Cf will be superior to ^{232}U as a source of radioactive recoiled materials for long-term storage of secret information. The latter, however, has a much longer half-life (i.e., can be used longer) than ^{252}Cf and requires no shielding against neutrons. Many useful recoiled materials can also be obtained through various nuclear reactions that produce radioactive species having suitable half-lives.[1]

SECRET MARKING OF EQUIPMENT WITH RADIOACTIVE SCREWS

For secret marking of office, laboratory, and shop equipment having more than one replaceable screw, replacement of one (or more) of the screws in each piece of equipment by a radioactive one of an identical type and size is suggested. A thief normally would not change all the screws in his stolen equipment, unless he

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happens to have read this article. Marking equipment with invisible radioactive inks may be objected to on the ground that bodily contamination is possible unless the radioactivities have been "baked" on the surfaces. Visible markings can be destroyed by anyone skillful in the art.

Radioactive screws were produced by bombarding screws with fast neutrons from a ^{241}Am–^{242}Cm–Be source[5] which emitted 4.6×10^7 unmoderated $n/(cm^2)(sec)$. An irradiation of brass, steel, and stainless-steel screws (size 6–32 by 1 cm) for 8 days produced, respectively, 870 dpm ^{65}Zn, 1.5×10^3 dpm ^{54}Mn, and 1.5×10^4 dpm ^{58}Co. These activities were below the detection levels of the conventional survey meters but were measurable by a gamma-ray spectrometer using a NaI(Tl) crystal as the detector.

A Cockcroft–Walton accelerator capable of generating 14-Mev neutrons with a flux of 10^9 $n/(cm^2)(sec)$ can produce approximately these same activities in a 1-hr irradiation. However, if a large number of screws are to be irradiated, a steady-state irradiation with the isotopic neutron source will be more desirable. Because the half-lives of ^{65}Zn, ^{54}Mn, and, particularly, ^{58}Co are rather short, periodic replacement by screws freshly made radioactive will be necessary.

SECRET MARKING OF HYDROGENOUS SURFACES WITH TRITIUM

The feasibility of using tritium for positive identification of objects that have hydrogen atoms on the surfaces has been studied. Tritium gas was allowed to exchange with the hydrogen atoms on the surfaces of papers, cloths, plastics, and woods. The existence of the invisible tritium marking was again revealed by autoradiography.

Various outgassed objects were exposed to 31 mc of tritium gas at 0.67 mm Hg and 24°C for 1 hr in a vacuum system. The objects included wooden sticks, tracing cloth, polymerized methyl methacrylate sheets, polyethylene foils, parchments, sheepskins, ledger sheets, check stubs, photographic copies, bank drafts, U. S. dollar bills, microfilms, tracing vellums, glossy prints, postage stamps, and bond, onionskin, and white starch papers. The tritiated samples were outgassed for several days and then kept in a dry atmosphere to minimize loss of tritium through exchange with the water molecules in the air. Autoradiography with the Ilford X-ray films showed that the tracing vellum and onionskin papers had relatively little tritium, the polyethylene foils had an undetectable amount, and all the rest contained an abundance of tritium.

The rates of loss of tritium from tritiated papers to a dry atmosphere and to an open atmosphere were studied. Strips of papers (Whatman filter papers No. 42, sheepskin parchments, Hammermill ledger sheets, and white starch papers), 1 by 4.5 cm, were exposed to 8.3 mc of tritium at 0.18 mm Hg and 24°C for 5 days or to 16 mc of tritium at 0.34 mm Hg for 24 hr. After being outgassed for several days, paper of each type was divided into two groups, one being kept in a desiccator with anhydrous $CaSO_4$ and the other in a hood with a constant air flow at an average humidity of 10%. Each strip of paper was counted by Tri-Carb liquid scintillation counter in a solution* of 5 g of PPO and 0.1 g of dimethyl POPOP per liter of toluene. Figures 3 and 4 show the relative decrease of tritium activities on the papers as a function of time. Tritium on papers exchanged rather rapidly with the moisture in the air.

Tritium gas also exchanges rapidly with hydrogen on papers. Two groups of sheepskin parchment strips were exposed to 30 mc of tritium at 0.64 mm Hg, one group for 1 hr and the other for 3.5 days. The ratio of the tritium activities of the 1-hr to the 3.5-day exposures on these paper strips was 0.66.

The present technique of secret marking with tritium has the feature that no chemical alteration ever takes place on the marked objects. Tritium has a half-life of about 12 years. The rate of loss of tritium in a dry sealed atmosphere is slow. In addition to autoradiography, disposable plastic scintillators can be used for nondestructive detection of tritium on the marked objects. However, the tritiated objects must be handled carefully so as to minimize tritium contamination through any prolonged inhalation or bodily contacts.

A SAFE-LOCKING SYSTEM BASED ON GAMMA-RAY SPECTROMETRY

In spectrometry, an intensity distribution of radiation energies, or some function of energies, is obtained which is characteristic of the substance(s) present in the sample. The intensity is a function of the amount of the substance(s) responsible for the characteristic radiations. In principle, a lock system may be envisioned which opens only with a key possessing the

*PPO = 2,5-diphenyloxazole; POPOP = 1,4-*bis*-2-(phenyloxazolyl)-benzene.—The Editor

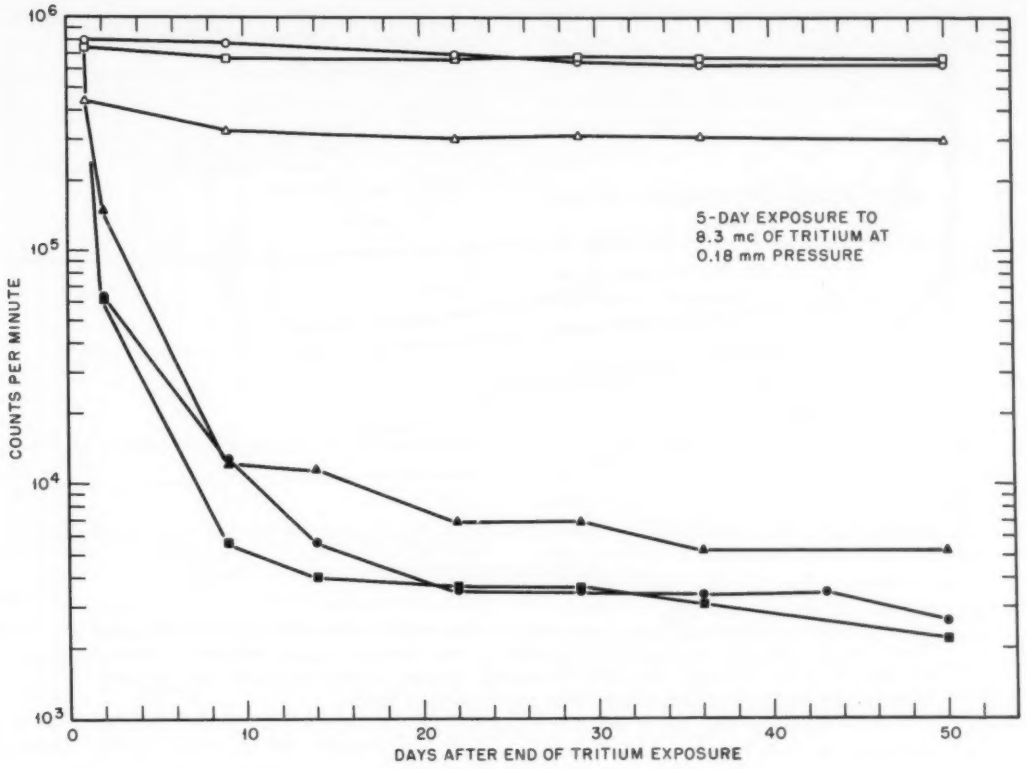

Fig. 3 Tritium activities on different paper strips as a function of the days after exposure. The papers were stored in either an open atmosphere (indicated by solid symbols) or a dry atmosphere (indicated by open symbols). Triangles: sheepskin parchment; squares: Hammermill ledger sheets; circles: white starch paper.

right combination of the preselected radiations and their associated intensities.[6]

The feasibility of devising a lock system using gamma-ray spectrometry has been studied. The lock consisted of two shielded NaI(Tl) detectors, a fast-slow coincidence spectrometer, a count-rate meter, a d-c voltage analyzer, and a relay locking mechanism. Figure 5 is a schematic diagram of the electronic units. The key was a radioisotope that emitted coincidence gamma rays. The rate meter converted the count rates from the coincidence circuit into output voltages. The voltage analyzer sorted out the voltages that fell within the limits set by the upper and lower windows. When the count rate matched the preset value, the output signal of the voltage analyzer closed the relay, and the lock opened. Forgery of the count rates by varying the counting geometries of the key was prevented by a sliding door (Fig. 6). The electronic equipment was concealed but accessible for repair. After the lock opened, the key was recovered by opening the sliding plate.

A margin of ±10% of a preset count rate, within which the relay would close, results in 32 distinguishable count-rate increments in the range 100 to 50,000 cpm. With an energy resolution of 10% for the detectors, there are 16 useful radioisotopes[7] that have half-lives between 1 and 10^6 years with gamma-gamma coincidences and gamma energies greater than 200 kev. There are, then, 16 X 32—or 512—combinations of radiation energies and intensities that are possible. The present counting technique accepted only the coincidence gamma radiations which were selected by the two single-channel analyzers and whose arrivals fell within the 50-nsec resolving time of the fast coincidence circuit. Incidental counting of similar gamma and Compton-scattered radiations was possible. For example, a ^{207}Bi source 20 times, or a ^{60}Co source 8 times, the strength of a ^{44}Ti source gave the

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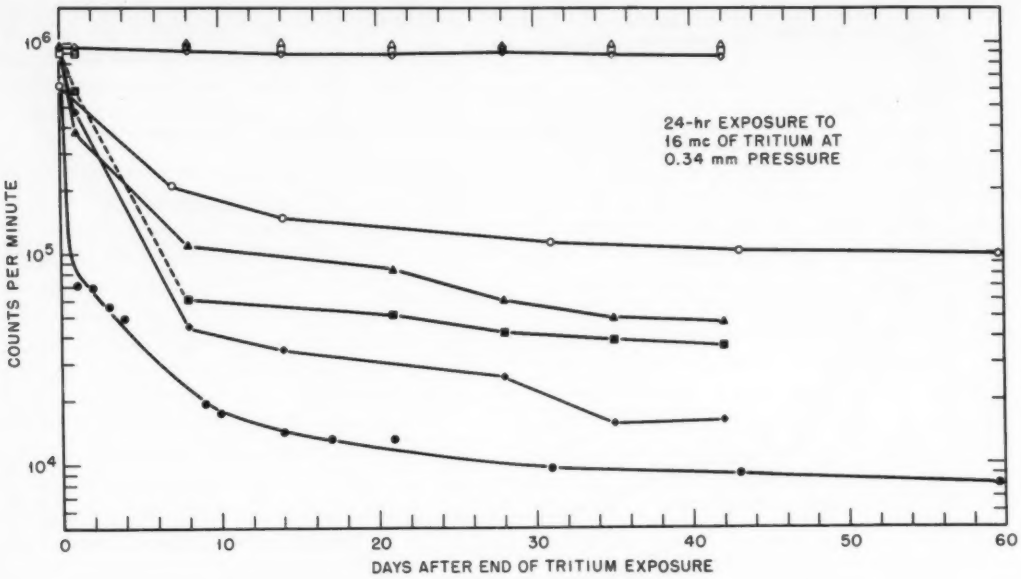

Fig. 4 Tritium activities on different paper strips as a function of the days after exposure. The papers were stored in either an open atmosphere (indicated by solid symbols) or a dry atmosphere (indicated by open symbols). Triangles: sheepskin parchment; squares: Hammermill ledger sheets; circles: Whatman filter paper No. 42; diamonds: Strathmore bond (25% cotton).

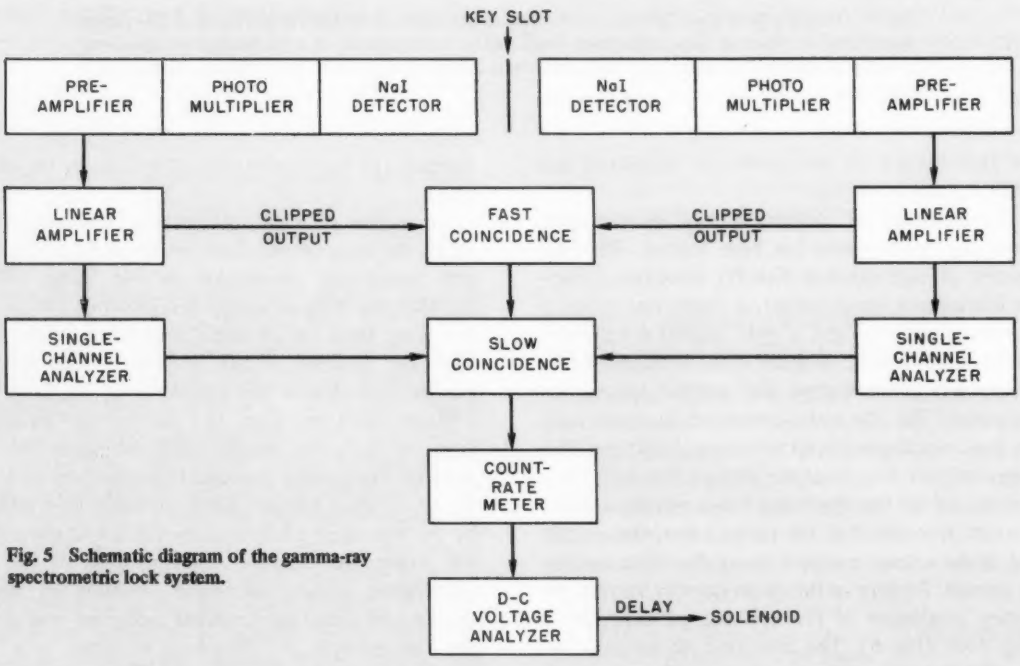

Fig. 5 Schematic diagram of the gamma-ray spectrometric lock system.

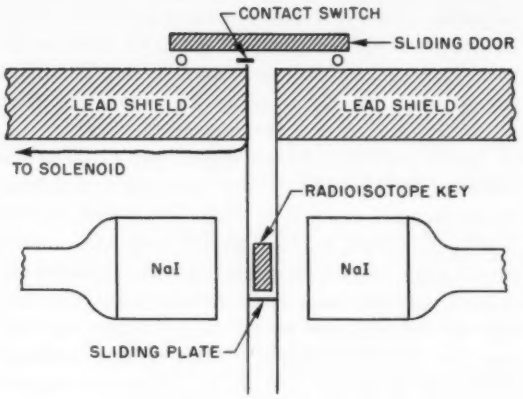

Fig. 6 Physical arrangement of the gamma-ray spectrometric keylock system.

same coincidence count rate as the latter when the 511- and 1156-kev gamma rays were accepted. The number of *unique* combinations of radiation energies and intensities is less than the expected 512 for this counting method.

Instead of counting coincidence gamma rays, a much better method is to define the shape of a selected photopeak with a three-channel analyzer and three rate discriminators. Three points can be shown to be necessary and sufficient to define uniquely a characteristic photopeak.[6] If energy and intensity resolutions are set at 10% each and the radioisotopes are limited to those having half-lives between 1 and 10^6 years with gamma energies[7] above 100 kev, there are 705 unique combinations of radiation energies and intensities in the range 100 to 500,000 cpm.

A timed lock with ^{198}Au was simulated, and only the 412-kev gamma radiations were counted. The key was left in the lock. The d-c voltage analyzer was set to accept the count rate predicted for the time at which the lock was to open. Radioisotopes can thus serve as useful silent timers.

The basic principle of this keylock system may be adapted to the many other types of spectrometries now available. In general, the lock consists of a radiation source and three intensity discriminators. The key is any substance that gives the selected characteristic spectral lines. When necessary, the key may be replaced by the same substance of different concentrations or by a different substance compatible with the spectrometry used. If the type of spectrometry to be used is kept secret, forgery will be quite difficult, since there are so many possible choices of chemical elements, compounds, and radioisotopes to be used for the keys. For practical purposes, the liquid solution or radioisotope keys do not have to be carried by anyone. The keys can be made up when needed and disposed of secretly.

DOOR INTRUSION DETECTION

As an extension of the previous system, a monitoring system for the detection of door intrusion[8] was developed. On a vertical door frame, 0.8 mc of ^{57}Co was mounted 85 cm from the floor, and a NaI(Tl) crystal (3.8 cm in diameter and 3.8 cm long) with photomultiplier and preamplifier was placed across the door at 90 cm from the ^{57}Co. The detector was equipped with a linear amplifier, a single-channel analyzer, a count-rate meter, a d-c voltage analyzer, and an alarm relay system. The single-channel analyzer accepted the 123-kev gamma radiations of ^{57}Co, and the rate meter converted the count rates into output voltages. Again, the d-c voltage analyzer sorted out the output voltages that fell within the limits set by the upper and lower windows. The relay system remained open at a preset count rate. As soon as the count rate received by the rate meter decreased or increased by 5%, the output signal of the voltage analyzer closed the relay which, in turn, sent out an alarm. The response took only a second. The alarm continued even when the count rate returned to the preset value. A 60-kg person walking slower than 60 cm/sec through the door would trigger the alarm. Power failure of the counting system also would close the relay and send out an alarm.

The thought occurred that perhaps the 1.1- to 1.3-Mev gamma radiations of ^{60}Co could differentiate between human bodies and steel chassis, as the energetic gamma rays might be attenuated more by steel (high atomic number) than by the elements of lower atomic numbers in human bodies. However, a hip of 30-cm thickness attenuated 80% of the ^{60}Co photopeak radiations, whereas a typical electronic unit with steel chassis and vacuum-tube circuitries attenuated only 61%. A person moving faster than 60 cm/sec through the door would escape detection by ^{60}Co.

For added effectiveness in detection, another detector was mounted on top of the door, 220 cm from the floor, and another ^{57}Co source was placed underneath the threshold, thus forming multiple radiation paths. The matched outputs of these two detectors were added to the single-channel analyzer. Anyone crawling through the door was easily detected by this system.

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In another experiment, for door security, 0.8 μc of ^{54}Mn was mounted on the inside of the swung-out edge of the door. With the door closed, the ^{54}Mn was 2 cm from the NaI crystal face. The analyzers' windows were set for the 840-kev gamma radiations and activities of the ^{54}Mn. When the door opened to a clearance of 2 cm, the alarm sounded.

The radioactive source may contain a mixture of isotopes which emit different gamma radiations to thwart would-be intruders. The use of an upper window in the voltage analyzer makes it impossible to duplicate the preset count rate with an intense radioactive source of high gamma energies and would trigger the alarm. Nuclear radiation detectors with little or no energy resolution, such as Geiger-Müller tubes, may be used in place of the expensive gamma-ray spectrometer. However, the degree of security will then be reduced, since many other radioisotopes could duplicate the preset count rate of the system.

The present system can be completely concealed, and this is an advantage over other intrusion-detection systems that use visible radiations. The concrete walls and the steel door frames also provide shieldings of the nuclear radiations, so that the radiation sources are difficult to detect by the intruders.

DISCUSSION

To utilize the invisible and characteristic nuclear radiations for security and law enforcement is potentially feasible in the few examples studied, where an element of secrecy is desirable. The present series of studies has not exhausted all the potential uses of radioisotopes for this purpose. Another possibility is a short-range secret communication using the gamma rays of ^{60}Co or ^{226}Ra (in equilibrium with its decay daughters) as the invisible transmitting signals. These gamma radiations have relatively low mass-attenuation coefficients and at least 30% can penetrate through 15 cm of concrete.[3] Thus a room-to-room secret communication through the common wall, with a gamma-ray detector as the signal receiver, can be envisaged. By moving the gamma emitter in and out of a lead shield, a dot-dash pattern of message can be sent which may be recorded automatically by a receiver. The system can be concealed almost completely.

The rapid exchange of tritium with hydrogen on surfaces offers a fast method of secretly marking and subsequent positively identifying a group of objects having hydrogenous surfaces. For example, a quick dispersion of a safe dosage of tritium gas into a case of canned goods will secretly mark the paper labels on the cans. However, for obvious reasons the use of tritium gas to mark quickly a group of persons for later identification is not to be recommended.

Secret and indiscriminate applications of detectable amounts of radioactivity near materials and instruments that are sensitive to nuclear radiations, such as X-ray films and nuclear particle counters, require careful considerations. In situations where radioactivity is used routinely, techniques such as those described may be objectionable to the experimenters and undesirable for the security, since a routine radiation survey will reveal the existence of the secretly planted radioactivities.

The proper use of the presently developed techniques and systems should produce no health hazards and no property damages. But, accidental ingestion and uncontrolled dispersion of the radioactive labels should, of course, be avoided. The pollution problems when using tritium to mark numerous surfaces for identification must be considered carefully.

The high costs of nuclear instruments at present make the two monitoring systems under study rather unattractive for general uses. However, if secrecy is the only important factor, the price of security may be willingly paid. (FEM)

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RADIOISOTOPE PRODUCTION IN PAKISTAN

By Abdur Rahman, Abdul Majeed, and Md. Matiur Rahman*

The Pakistan Institute of Nuclear Science and Technology (PINSTECH) has a 5-Mw swimming-pool type research reactor (PARR), which went critical in December 1965. PINSTECH has been designed to serve as a national center for the entire country to provide facilities for advanced training and research in practically all the important fields concerned with the development of atomic energy in Pakistan. The reactor time for isotope production is thus limited, and it may not be feasible to meet all of Pakistan's isotope requirements from PINSTECH.

Radioisotopes were being applied in medicine, agriculture, and scientific research 5 to 6 years earlier, requirements being met entirely by imports. In 1965–1966 as many as 52 different radioisotopes were being used, excluding large radiation sources; those most frequently used were ^{131}I, ^{32}P, ^{51}Cr, ^{55}Fe, ^{35}S, and ^{85}Kr. The users were 80% in medicine, 19% in scientific research, and 1% in industry. The initial program called for production and processing of ^{131}I, ^{32}P, and ^{35}S on a plant scale; of ^{51}Cr, ^{198}Au, ^{59}Fe, and ^{65}Zn in batch lots; and of isotopes not requiring processing, such as ^{192}Ir, ^{198}Au (implantation seeds), ^{24}Na, and ^{42}K.

Besides other facilities such as beam tubes, PARR has one 2.5-in.-diameter vertical tube for in-core irradiation, three 2-in.-diameter pneumatic rabbit tubes, and a graphite thermal column. The PARR core being flexible, its configuration can be rearranged so that one or more vacant positions are available for irradiation samples contained in dummy elements. The average thermal-neutron flux in the core at maximum power load is about 3×10^{13} n/(cm^2)(sec), and the cadmium ratio is about 8. The neutron flux in the rabbit tubes is about 2×10^{13} n/(cm^2)(sec). In the vertical tube the maximum neutron flux is 5×10^{13} n/(cm^2)(sec) at a height of 27 cm from the bottom.

The vertical tube and the pneumatic rabbits have been used for irradiation. For longer irradiations, standard NRX cold-welded capsules are suitable. The capsules are loaded in open-ended aluminum sample holders (Fig. 1), which are placed in holes at the periphery of the reactor core (Fig. 2). After irradiation, the capsule holders are transferred to a steel tray affixed to the pool wall some 7 ft from the surface of the water in the pool. A push-pull mechanism extracts the capsules, which are stored on the tray until required or until the ^{24}Na induced in the aluminum capsules has decayed. Facilities are also provided for underwater transfer of irradiated samples to a dry hot cell equipped with remote manipulators, viewing window, and an overhead 1-ton crane.

Processing cells are provided with necessary shielding and standard equipment for handling radioactive materials, waste disposal, and personnel safety.

Though irradiation of samples started at PARR soon after it went critical, the first irradiation† for production of an isotope for clinical use inside the country was in 1967 for ^{24}Na, followed by ^{42}K. These are being regularly supplied at millicurie levels to the Cholera Research Laboratory at Dacca, East Pakistan, 1500 miles away. Procedures have been standardized for making ^{198}Au available to local hospitals and the short-lived ^{24}Na, ^{42}K, ^{82}Br, ^{121}Sn, ^{140}La, and others to educational institutions throughout the country.

(MG)

(FIGURES 1 AND 2 ARE ON PAGE 342.)

*Isotope Production Division, Pakistan Institute of Nuclear Science and Technology, Nilore, Islamabad.

†Md. M. Rahman, A. Rahman, and M. A. Mubarak, Production of the First Commercial Batch of Radioactive Isotopes at PINSTECH, Pakistani Report PINSTECH/R1PD-1 (1968).

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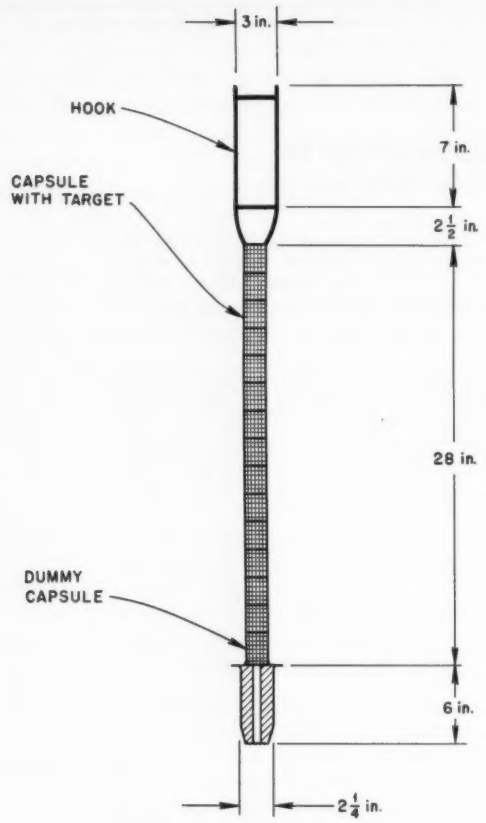

Fig. 1 Capsule holder for in-core irradiation.

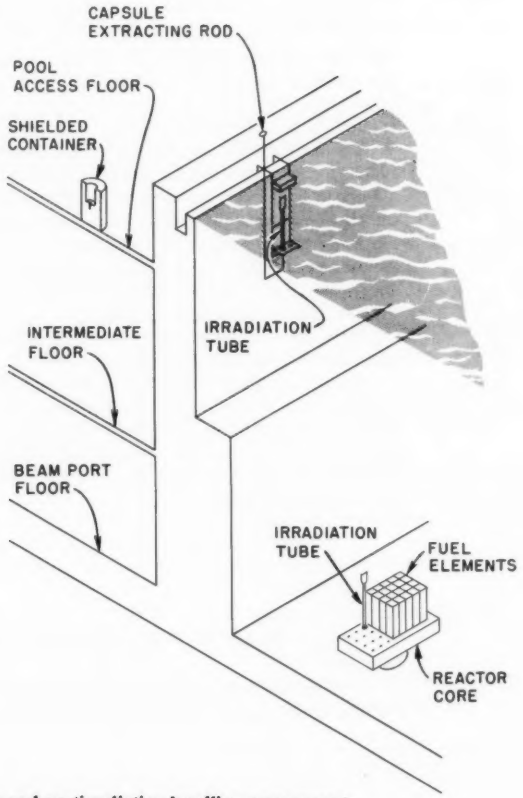

Fig. 2 Simplified general plan for irradiation and postirradiation handling arrangements.

Section

III

Isotopes and Radiation Technology

Applications in Life Sciences

New Techniques To Study Nutrient Relations in Plants*

By G. D. Bowen and A. D. Rovirat

Abstract: *A new technique for easy examination of variations of many properties along roots involves the combined use of the radiochromatogram scanner and tracer experiments. This technique has also been used for studying the effects of specific treatments on the activity of different parts of roots. Large differences along roots demonstrate the dangers in interpreting data obtained by averaging the results from an entire root. The basic scanning technique used can be applied, with minor modifications, to many types of biological material.*

Radioisotopes have been used extensively in determining the uptake of nutrients by plant roots, but most uptake studies use the entire root system without considering variations along roots. Very few detailed studies have been made on differences in uptake, loss, and biochemical incorporation of nutrients at different sites along plant roots. Selective feeding of parts of roots with isotopes[1,2] and counting of individual segments of roots, cut either before or after exposure to radioisotopes,[3,4] are laborious methods when replication is required. The need for techniques that are accurate, quantitative, and simple enough to permit the replication needed for biological material has been met by the new methods described here.

In our studies, some of which are indicated in this article, we have used radioactive phosphate, sulfate, chloride, and carbon dioxide to study environmental factors affecting uptake, loss, and biochemical incorporation of the labeled compounds along roots. The results have provided information on (1) root characteristics important in nutrient uptake from soils; (2) models of ion movement from soils to roots; (3) formulation of questions on, for example, the differential control of ion uptake and use along plant roots; and (4) the sites along roots most likely to be important in release of organic compounds that stimulate microbial growth around roots.

BASIC METHOD

Radioisotopes are absorbed by roots from radioactive solutions, and then the sites where radioactivity occurs along roots are recorded by mounting the roots on 4-cm-wide chromatography paper (Whatman No. 1), which is then passed through a gas-flow radiochromatogram scanner (Fig. 1). For phosphate and chloride studies, where the emitted radioactivity is strong, the roots are attached to the paper by covering them with cellulose tape using an assembly (Fig. 2) devised by C. P. Eickhoff, Division of Soils, Commonwealth Scientific and Industrial Research Organization. With weak emitters, such as ^{35}S and ^{14}C, the paper strip is treated with a contact adhesive, and the roots are covered with thin polyethylene sheet (1.1 mg/cm^2). This decreases the recorded radioactivity from ^{35}S by 26%, compared with 40% if cellulose tape is used. Drying of the mounted roots at 40 to 50°C for 30 min prevents redistribution of the isotopes after mounting. With optimal settings of collimator width on the detectors, scanning speed, and of integration time for the scale at which the sample is being scanned, the resolution of

*Reprinted, with minor editorial changes, from *Atomic Energy in Australia*, 12(1): 2-7 (1969), by permission of the publisher.

†Division of Soils, Commonwealth Scientific and Industrial Research Organization, Glen Osmond, South Australia.

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this method is better than 2 mm, and radioactivity at any portion of the root is directly proportional to the area under the corresponding part of the scan, provided there is uniform self-absorption of radiation along the root. For the 3-week *Pinus radiata* and the 2-week wheat seedlings used in our experiments, this condition is met.

By suitable experimental design, the basic method, described elsewhere[5] in more detail, provided the types of information described below.

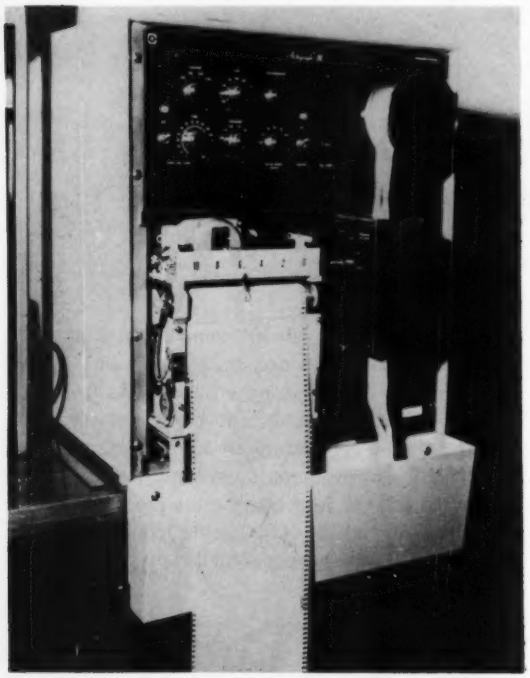

Fig. 1 Radiochromatogram scanner scanning pine seedlings.

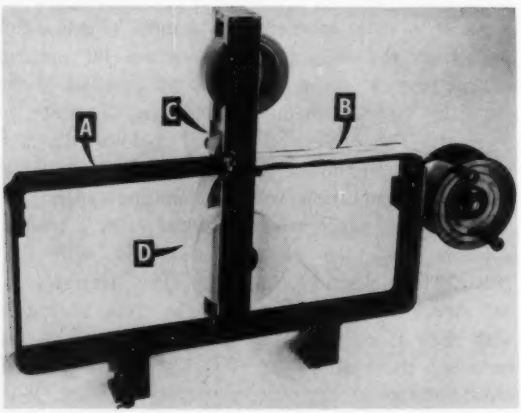

Fig. 2 Instrument used for fixing labeled wheat roots (*A*) and tops (*B*) between cellulose tape (*C*) and filter paper strip (*D*).

SITES OF CHLORIDE, SULFATE, AND PHOSPHATE UPTAKE

When the uptake time from labeled solutions was restricted to 15 min followed by a 5-min washing to remove nonabsorbed tracer, only 5 to 10% of the absorbed radioactivity was translocated to the tops of experimental plants; this and other studies have shown that in 15 min the sites of accumulation are essentially those of uptake for wheat roots and pine-seedling roots.

For the composite diagram[6] shown in Fig. 3, wheat was grown in soil for 14 days, and the roots were carefully removed by washing away the soil prior to insertion in nutrient solution containing radioactive

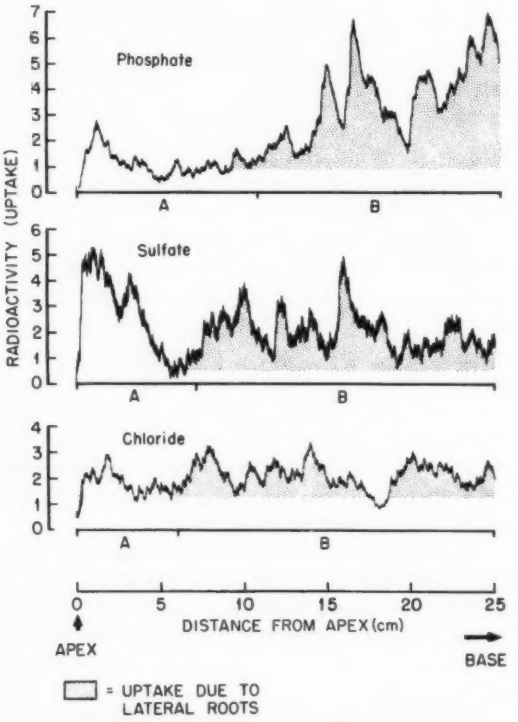

Fig. 3 Phosphate, sulfate, and chloride uptake along the apical 25 cm of main and lateral roots of soil-grown wheat. (From Ref. 6.)

chloride, sulfate, or phosphate. With all three anions, there was a high uptake in the apical 4 cm. With sulfate, there was no uptake behind this zone except where lateral roots occurred. Phosphate and chloride uptakes in the prelateral root zone declined to about 25% of that in the apical region, but there was dramatic increase in uptake of sulfate and phosphate with the onset of laterals, and the combined uptake by the lateral roots far exceeded that of the main root. In both cases the lateral roots accounted for more than 70% of the anion uptake of the entire root system. This very high uptake by the lateral roots indicates the importance of placement of fertilizers into that zone of the soil which will be explored by lateral roots.

The results raise basic questions on the factors affecting lateral root production in soil, control of uptake of ions at different parts of roots, and the differences in this control with different ions. Differences between ions in uptake patterns along the apical regions of roots have also been described.[7]

These experiments have been extended to study the effects of environment—e.g., soil temperature, nutrient levels in soil, and soil sterilization—on the patterns of uptake along wheat and pine roots grown in soil and in nutrient solution.

ZONES OF PHOSPHATE TRANSLOCATION

The uptake and translocation of phosphate from the roots to the tops of plants were studied by treating the roots with ^{32}P–phosphate in calcium sulfate for 15 min, removing some of the plants for scanning, and transferring the remainder to ^{31}P–phosphate in calcium sulfate for 3.5 hr to allow translocation of the radioactive phosphate from the roots to the tops. Differences in radioactivity between the initial series of plants and those allowed to translocate for 3.75 hr indicated the extent of translocation from various parts of the roots in that time. The loss of phosphate to solutions surrounding the roots was slight and did not affect the results.

Scans (Fig. 4) of typical roots and tops after a 15–min uptake and translocation time showed high uptake in the apical 2 cm of the root followed by a decrease until the appearance of lateral roots, where uptake was very high. After an uptake and translocation time of 3.75 hr, the radioactivity of the apex decreased to 60% of that after 15 min, while in the mid and lateral portions of the roots, almost all the radioactive phosphate had been translocated to the tops. These results indicate very clearly that the mid and lateral root regions are very active in taking up phosphate, which is then translocated to the tops, while in the high–uptake zone of the apical root portion, a high proportion of this phosphate is retained and probably used by the dividing and elongating cells. Similar results have been obtained with pine seedlings.

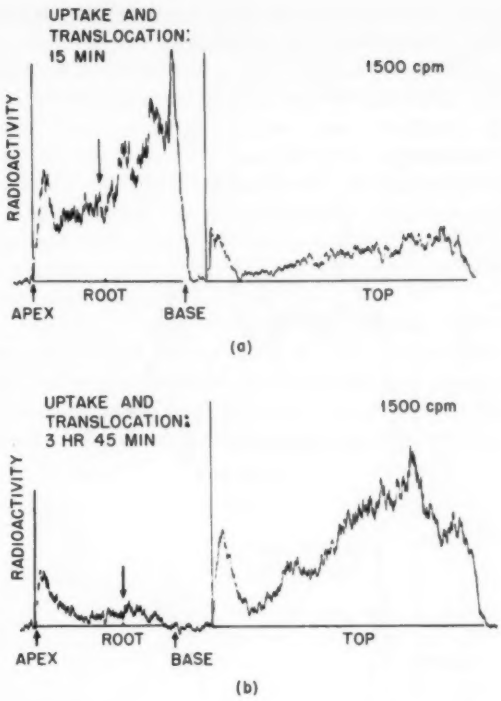

Fig. 4 Translocation of phosphate from the roots to the tops of wheat. Only one of the five roots contributing to the translocation to the top of each plant is shown. The beginning of the lateral root zone is shown by arrows pointing downward.

BIOCHEMICAL INCORPORATION

Some of the biochemical differences along roots can be demonstrated by scanning the root before and after extraction by solvents for particular groups of compounds. The root "skeleton" must be left intact by the extraction method, but extraction of the compounds from the plant species used must be efficient. In this type of study, it is more satisfactory experimentally to perform the scanning sequences on the same roots, and the roots are mounted between thin polyethylene sheets before being placed on the chromatogram paper strip. The polyethylene prevents

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drying out of the roots and enables easy removal of them for the next extraction procedure.

Figure 5 was made from microbiologically sterile roots of pine seedlings grown in a high-phosphate plant nutrient solution for 3 weeks and then fed ^{32}P in potassium dihydrogen phosphate for 15 min.

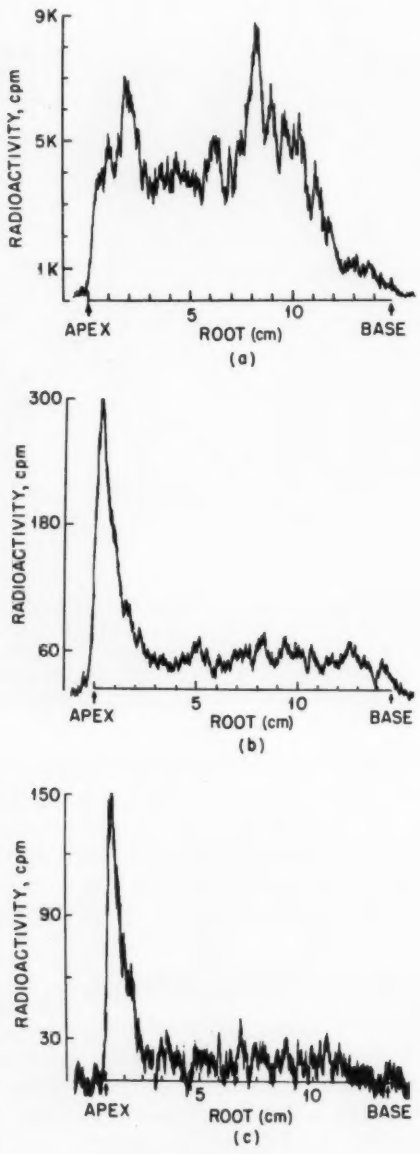

Fig. 5 Incorporation of ^{32}P-phosphate along pine roots. (*a*) Total ^{32}P in roots. (*b*) ^{32}P distribution after removal of inorganic phosphate and esters by repeated freezing and thawing in 0.1*N* $HClO_4$. (*c*) ^{32}P (in phosphoprotein and phospholipid) after removal of nucleic acids by the Ogur–Rosen method.

The roots were washed free of nonabsorbed ^{32}P and 2 hr later were scanned ("total phosphate," Fig. 5*a*). Then the inorganic orthophosphate, sugar phosphate, nucleotides, and other easily acid-soluble compounds were removed into 0.1*N* perchloric acid with repeated freezing in solid carbon dioxide and thawing. The subsequent scan (Fig. 5*b*) gave the distribution of labeled nucleic acids, phospholipids, and phosphoproteins along the root. The nucleic acids were then removed by the Ogur–Rosen method,[8] and the roots were scanned again (Fig. 5*c*). Calculation of differences for various parts of the roots between successive scans showed large differences along the roots in the relative incorporation of phosphate into the three phosphate fractions. This information was supplementary to that for sites of translocation of phosphate from roots to tops. The technique and incorporation data for different segments of pine seedlings grown under high- and low-phosphate conditions were given in detail by Bowen.[9]

INORGANIC ION LOSS FROM ROOTS

A detailed understanding of ion relations of roots includes the study not only of ion-uptake sites ("influx") but also of loss sites ("efflux"). The sites of chloride loss from pine roots[10] were obtained from *P. radiata* seedling roots grown aseptically in plant nutrient solution containing ^{36}Cl-chloride for 4 weeks and then washed in running tap water for 10 min. The sites of efflux were determined by placing the roots between strips of Whatman No. 1 chromatography paper moistened evenly by spraying with plant nutrient solution containing no chloride. The paper strips and roots were held between polyethylene-covered foam plastic on a hardboard base. The plants were placed in a moist chamber, and the loss of radioactive chloride from the roots was studied at intervals for 24 hr by periodically changing the papers, drying them, and measuring the radioactivity along the paper with the chromatogram scanner-recorder. The distribution of residual radioactive chloride in the root at the end of 24 hr was obtained by scanning.

Figure 6*a* shows that the apical portion of the pine root contained only about the level of chloride of the mid-root portion. The efflux of chloride along the root was not detectable after a 1-min contact between the roots and the paper. However, there was a low but uniform chloride loss along the entire length of the root; and, during the next 20 hr, the chloride loss from

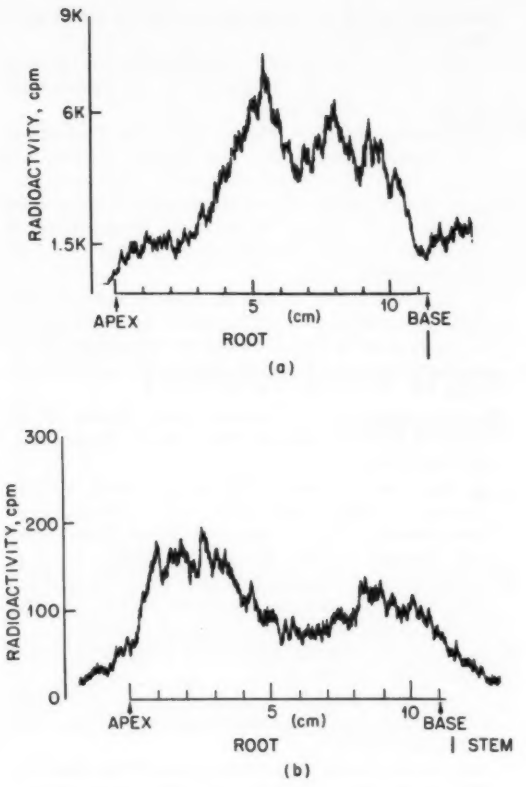

Fig. 6 Chloride loss from pine roots. (*a*) ^{36}Cl-chloride distribution in roots. (*b*) Loss of chloride over 20 hr from the same root.

the apical region was greater than that from the basal region (Fig. 6*b*) despite the much lower chloride content in the apical portion.

The relative loss from the apical portions of the root is quite different from that from other parts of the root, and, over a 24-hr period, it may be up to ~20% of the chloride in the apex. Obviously the study of efflux at different parts of the roots is necessary to the critical interpretation of many nutrition experiments.

EXUDATION OF ORGANIC COMPOUNDS FROM WHEAT ROOTS

Intact healthy plant roots exude organic compounds, but little is known of the exudation from different parts of the roots. Carbon-14 has been used to measure this exudation from individual roots,[11] and these studies have now been extended to study the leakage from different parts of the wheat roots using the filter-paper "sandwich" technique. Wheat plants were grown for several days with their roots between 4-cm-wide strips of Whatman No. 1 filter paper, wet with plant nutrient solution and contained in polyethylene tubing held together with slight pressure by foam-plastic-covered hardboard. The polyethylene tubing extended above this sandwich to enclose the tops and was sealed to provide a completely enclosed atmosphere into which $^{14}CO_2$ was injected at a suitable stage of growth.

The scans in Fig. 7 were obtained when a plant received a single pulse of 50 μc of $^{14}CO_2$ and was then allowed to grow between the filter paper for a further 48 hr. The position of the roots was marked on the paper. Then the radioactivity inside the root and that

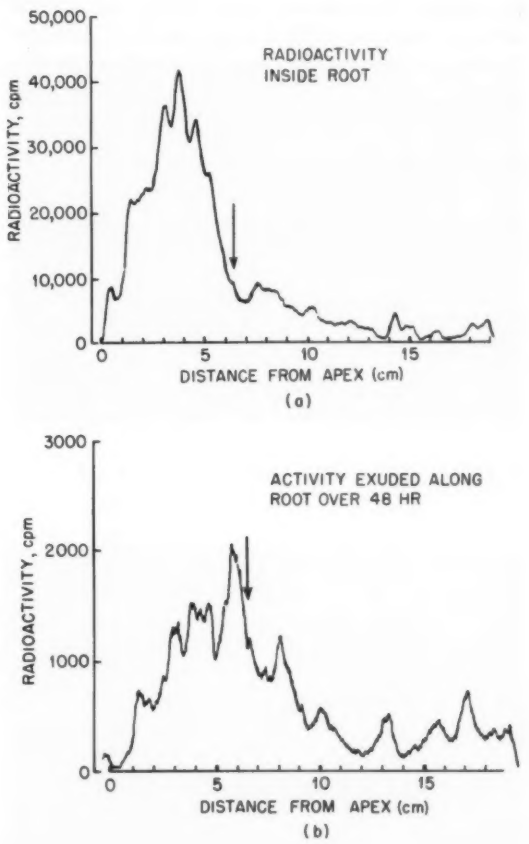

Fig. 7 (*a*) Distribution of ^{14}C along wheat roots 48 hr after labeling of tops with a pulse of 50 μc of $^{14}CO_2$. (*b*) Exudation of ^{14}C from wheat root (*a*). The arrows indicate position of root apex when $^{14}CO_2$ was administered to the tops.

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which exuded into the paper were measured and recorded by passing the roots and the paper through the chromatogram scanner.

These scans show that most of the radioactivity accumulated in the apical 6 cm of the root that developed after administration of the $^{14}CO_2$ (Fig. 7*a*). The chart of the paper in contact with the root showed that most exudation occurred in the apical 9 cm of the root (Fig. 7*b*). Subsequent experiments indicated that less than 30% of this exudate from the apical region diffused away from the root, while more than 70% of that from the older parts of the root was diffusible.[12]

ACKNOWLEDGMENTS

The radiochromatogram scanner used in these studies was purchased from a grant made by the Rockefeller Foundation, New York, U.S.A. The technical assistance of M. B. Davies and A. E. Langham throughout these investigations is gratefully acknowledged. (MG)

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Section

IV

Isotopes and Radiation Technology

Safety

Tritium Loss from Tritium Self-Luminous Aircraft Exit Signs*

By R. G. Niemeyer

Abstract: *Tritium loss from self-luminous aircraft exit signs was studied. Although only about 2% of the tritium in the light sources was in the oxide form, tritium oxide was measured because the biological effect of the oxide is greater than that of elemental tritium. About 99% of the tritium released from a broken exit sign escapes in the first 1.75 hr. The permeation rate of tritium through the intact glass tubes is so low as to be inconsequential from the safety standpoint.*

Self-luminous aircraft exit signs (Fig. 1) contain a tritium-activated phosphor light source made by coating the inside of glass tubing with phosphor, loading the tube with tritium gas, and sealing the tube. The light from the source reflects from a white surface and illuminates the exit sign. Three exit signs and 10 light-source tubes (Fig. 2) were received from the manufacturer for testing. Light sources made at Oak Ridge National Laboratory (ORNL) were also tested; these sources were made, without phosphor, from tubing supplied by the manufacturer and tritium supplied by ORNL. Tests covered the total amount of tritium contained in the tubes, the rate of tritium leakage from a broken exit sign, the possibility of tritium reacting with the potting compound holding the tube in place, and the permeation rate of tritium through an intact tube. The ORNL sources were tested to determine the effect of aging on the amount of ^3H_2O collected in the tube.

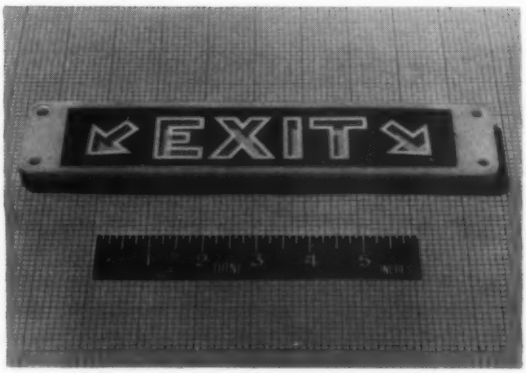

Fig. 1 Tritium self-luminous aircraft exit sign.

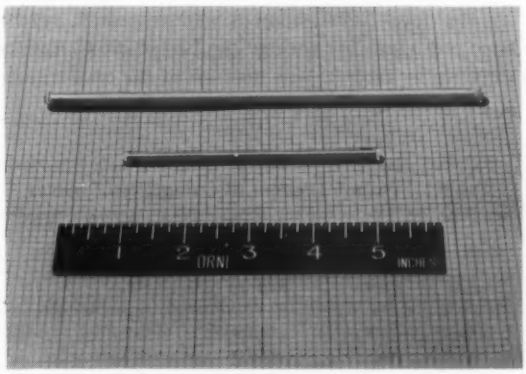

Fig. 2 Tritium-activated phosphor light-source tubes.

*A condensed version of: R. G. Niemeyer, Tritium Loss from Tritium Self-Luminous Aircraft Exit Signs, USAEC Report ORNL-TM-2539, Oak Ridge National Laboratory, Mar. 25, 1969.

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TEST EQUIPMENT

The test equipment used consisted of a carrier-gas system, a glass containment vessel, and a trapping system. The glass containment vessel had a mechanical device for breaking the tube containing the tritium. The trapping system consisted of five cold U-traps and one hot (700°C) copper oxide bed. The gas flowed through two cold traps to remove any ^3H_2O that may have been present, then it entered the hot copper oxide bed where the elemental tritium gas was converted to ^3H_2O. The tritium oxide was recovered in a series of three cold traps, and the exit gas passed through a backup water scrubber before going to the hot off-gas. Air flow rates were monitored with a calibrated rotameter. Tritium-gas samples of about 10 mc and 4 curies were released in the break-seal chamber to test the system. About 70% of the activity was recovered in the first 45 min, and about 99% was recovered in the first 4 hr when a dry air flow of 18 liters/hr was used.

TRITIUM CONTENT OF SOURCES

Four commercial sources were analyzed. Two were 2.4 years old, contained a nominal activity of 4.0 curies, and were made of 6-mm tubing, 7.375 in. long. The other two were 2.5 years old, contained a nominal activity of 1.85 curies, and were made of 5-mm tubing, 4.125 in. long. All four tubes had a 1-mm wall thickness. The sources were broken in the containment vessel, and an air flow of 2.7 changes per hour at 25°C was maintained for 6 hr and then at 700°C for 1 hr. Data are given in Table 1. In three cases, about 95% of the tritium given off during the 1 hr at 700°C was in the form of ^3H_2O; in the other case, about 85%. In all four samples, nearly all the elemental tritium was removed at 25°C.

The four ORNL sources were measured by the same procedure, but without the 1 hr at 700°C. Results are given in Table 2. Less than 1% of the total tritium was found in the backup water scrubbers.

Table 1 Tritium Content of Commercial Sources

Nominal activity, curies	Measured activity, curies	^3H_2O, %
4.0	4.09	1.6
4.0	4.49	1.3
1.85	1.73	6.8
1.85	1.74	2.7

Table 2 Tritium Content of ORNL Sources

Calculated activity, curies	Measured activity, curies	^3H_2O, %
4.0	3.82	1.1
3.5	3.29	1.5
3.0	2.82	2.1
1.0	0.94	1.6

TRITIUM RELEASE FROM A BROKEN EXIT SIGN

A 2.4-year-old exit sign that contained 3.1 curies of tritium at the time of manufacture was placed in the containment vessel, and both the sign and its tritium tube were broken. The gas was continuously collected for 212.25 hr, with the traps being replaced at the end of each collection period to obtain the tritium release rate. Results are given in Table 3. The first 1.75 hr accounted for 98.9% of the total tritium recovered. The percentage of tritium released as oxide in each collection period increased from 0.01% in the first 1.75 hr to 91.4% in the last 137 hr; however, the total tritium released was smaller with each successive period. A total of 2.14 curies was recovered.

Table 3 Tritium Released from Exit Sign

Gas collection period, hr	Tritium released, mc	Average release rate, mc/min
0.25	577.11	38.47
0.5	761.42	25.38
1.0	771.47	12.86
1.5	8.20	0.0911
17.0	7.45	0.0073
31.0	5.54	0.0029
24.0	2.43	0.0017
137.0	2.70	0.0003

REACTION BETWEEN TRITIUM GAS AND POTTING COMPOUND

The light-source assembly of an exit sign consists of an aluminum channel in which the tritium tube is held in place by a surrounding potting compound. A light-source assembly was removed from an exit sign and placed in the containment vessel. An air flow of

2.7 changes per hour was maintained for 48 hr to sweep out any tritium that might have been escaping from the assembly. The vessel was evacuated, and the light source was broken. The nominal 4 curies of tritium was allowed to react with the assembly for 11 days; then the tritium gas was collected continuously for 700 hr. Results are given in Table 4; some of the tritium may have been in the form of an organic species since the potting material is organic. A total of ~3.1 curies was recovered. The tritium oxide content was 3.1%, which indicates that some reaction had occurred between tritium gas and the potting material inasmuch as this is much more tritium oxide than was recovered from the broken sign in the previous experiment when the potting material was not soaked in tritium gas. Afterward, the source assembly was immersed in water for 30 days; an additional 35 mc of tritium was recovered.

Table 4 Tritium Released from Light-Source Assembly After Exposure to Tritium Gas for 11 Days

Gas collection period, hr	Tritium released, mc	^3H_2O, %	Average release rate, mc/hr
3.5	2993	0.9	885.0
26.0	58	91.4	2.2
331.5	6	83.3	0.018
24.0	1	90.0	0.042
315.0	9	97.8	0.029

PERMEATION OF TRITIUM THROUGH GLASS TUBES

Six intact commercial tritium light sources, i.e., tritium-filled glass tubes, were sealed in a small containment vessel, and tritium that permeated through the tubes was removed by an air flow. Results are given in Table 5. For the first samples, very long sampling periods were required to exceed the minimum quantitative level of detection (4000 dpm per milliliter of water). For samples 7 through 20, tritium was removed from the traps by rinsing with scintillation fluid rather than water; this reduced the minimum level of detection to about 20 dpm per milliliter of fluid. The Dry Ice traps were replaced with liquid-nitrogen traps to reduce the vapor pressure of ^3H_2O. The air sweep was replaced by a nitrogen-gas sweep to prevent condensation in the collection traps and to prevent oxidation of tritium. For samples 10 through 19, the atmosphere in the sealed vessel was changed from air to nitrogen, and the traps were changed back to Dry Ice. For sample 20, the atmosphere in the sealed vessel was changed back to air, the traps were changed back to liquid nitrogen, and nitrogen was used to sweep the sample out of the collection chamber. These changes, made to improve the analytical procedures and to obtain data on the factors that influence the amount of ^3H_2O trapped, are not believed to have affected the detection of the total amount of tritium in the samples. Of the total of 1.144 μc collected during 2463 hr, 0.598 μc, or 52.3%, was in the form of tritium oxide.

Table 5 Permeation of Tritium Through Glass Tubes

Sample number	Sampling duration, hr	Tritium permeation, pc/hr per tube	^3H_2O, %
1	72	113	7
2	143	30	50
3	190	190	3
4	239	32	16
5	336	32	19
6	402	21	10
7	242	84	92
8	169	229	82
9	191	173	79
10	191	61	93
11	24	103	69
12	24	193	61
13	24	20	75
14	72	26	65
15	24	47	53
16	24	24	29
17	24	15	47
18	24	47	28
19	24	18	28
20	24	20	25

The data in Table 5 can be expressed by the permeability velocity constant, K, using the relation

$$q = KAt\,(p_2 - p_1)/d$$

where q = total volume of tritium gas at STP, cm^3
t = time, sec
A = surface area, cm^2
p_2 = partial pressure of tritium inside the tube, cm Hg
p_1 = partial pressure of tritium outside the tube, cm Hg
d = wall thickness of tube, mm
K = permeability velocity constant, $(cm^3)(mm)/(cm^2)(sec)(cm\ Hg)$

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For one tube,

$q = 7.3 \times 10^{-8}$ cm^3
$t = 8.87 \times 10^6$ sec
$A = 59.0$ cm^2
$p_2 = 50.3$ cm Hg
$p_1 = 0$ cm Hg
$d = 1$ mm
$K = 2.77 \times 10^{-18}$ (cm^3)(mm)/(cm^2)(sec)(cm Hg)

If q is expressed as the total activity permeating the glass tube in curies, then K is 7.20×10^{-18} (curies)(mm)/(cm^2)(sec)(cm Hg). Although this value of K is very low, it is not inconsistent with the data of Norton[1] and Altemose.[2] Very large differences in the value of K occur for different glasses. Hydrogen is known to diffuse through glasses much slower than helium because it is influenced by the chemical bonding forces between the constituents of the glass. Soda–lime glass should exhibit a much lower permeation rate than a pure glass such as fused silica.

CONCLUSION

About 2% of the radioactivity in the light-source tubes was assumed to be in the form of tritium oxide. The probable origins of the tritium oxide are the original tritium bulk supply and the tritium reaction with residual oxygen or water adhering to the inside of the glass tube. The tritium oxide appeared in about the same amount both in freshly prepared tubes and in tubes about 2.5 years old. Tritium gas apparently does react slowly with the potting material holding the light source in place inside the exit sign. The permeation rate of tritium through the glass tubes used in these devices is so low that it is considered inconsequential from the safety standpoint. (FEM)

REFERENCES

1. F. J. Norton, Helium Diffusion Through Glass, *J. Amer. Ceram. Soc.*, **36**(3): 90-96 (1953).
2. V. O. Altemose, Helium Diffusion Through Glass, *J. Appl. Phys.*, **32**(7): 1309-1316 (1961).

Section

V

Isotopes and Radiation Technology

Miscellaneous

Availability of Isotopes and Services

RADIOISOTOPE SOURCES

Strontium-90

Isotopes, a company in Palo Alto, Calif., has announced a new irradiator in which two ^{90}Sr–^{90}Y sources, in an opposed geometry, achieve dose rates up to 15 thousand rads/hr. Dose uniformity is better than ±5% in two dimensions and is ±10% for samples less than 200 mg/cm^2 thick. The dose delivered is highly reproducible and depends only on the timing. The 28-year half-life of the source obviates frequent correction for decay. Radiation effects produced are equivalent to those produced by ^{60}Co.

The sample-holder volume is 1.2 cm^3. A slide arrangement places the sample in position to be irradiated in the chamber, an aluminum housing with dimensions of 6 by 6 by 6 in. and weighing only 20 lb. The external dose rate is less than 3 mrads/hr at 1 ft. The device was developed as a calibration source for thermoluminescent dosimeters but will find wide use in biology and solid-state physics as a high-dose-rate irradiator for small samples. The complete price is less than $800.

RADIOISOTOPES

High-Purity ^{242}Pu, ^{233}U, and ^{234}U

The U. S. Atomic Energy Commission (AEC) is making available for sale, for the first time, high-purity ^{242}Pu, ^{233}U, and ^{234}U, enriched by the electromagnetic process at Oak Ridge National Laboratory, Oak Ridge, Tenn., and by radioactive disintegration processes at Mound Laboratory, Miamisburg, Ohio. These first two isotopes mentioned are used by nuclear fuel processors as tracers for radiometric and mass-spectrometric measurements and analyses of yields in reactions involving plutonium and uranium. They are also useful as alpha standards for instrument calibration. The third is used for the manufacture of fission chambers to detect in-core thermal-neutron fluxes in nuclear reactors. Prices, per milligram, are $15.89 for ^{242}Pu with an isotopic purity of 90% or greater and $2.58 and $8.32, respectively, for ^{233}U and ^{234}U with an isotopic purity of 99.9% or greater. Each material will be supplied as an oxide. Other high-purity isotopes of plutonium and uranium as well as of thorium will be offered for sale as they become available.

Persons purchasing these isotopes for use in the United States must have an appropriate license issued by either the AEC or a state to which the AEC has transferred certain regulatory authority over radioactive materials. Domestic requests to purchase should be sent to: Isotopes Sales Department, Oak Ridge National Laboratory, P. O. Box X, Building 3037, Oak Ridge, Tenn. 37830. Requests for distribution abroad should be directed to the Division of International Affairs, USAEC, Washington, D. C. 20545.

Cobalt-60

The AEC is offering to make available, without charge, high-specific-activity ^{60}Co—more than 200 curies/g—to U. S. firms or organizations willing to undertake research and development work with their own funds on heat-source applications. Title to the ^{60}Co would remain in the AEC. Participants will be initially selected on the basis of a Request for Proposals to be issued, but unsolicited proposals will be considered later. Proposed programs of interest to AEC will be given preference. The offer is also available to other government agencies for heat-source applications research and development other than on space heat-

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source conversion systems. Requests from foreign organizations will be considered to the extent that adequate supplies remain available to accommodate domestic requests.

Contractors using the government-supplied material will be expected to provide their research and development results to AEC for such use and dissemination as AEC shall determine to be in the best interest of advancing this area of technology. The program will complement on-going and anticipated AEC heat-source research and development programs.

Further information on the program may be obtained from the Director, Division of Isotopes Development, USAEC, Washington, D. C. 20545.

Cesium-137

The AEC has canceled a proposal, announced in June 1968, to increase ^{137}Cs prices. Present prices, which have been in effect since late 1965, are:

Quantity purchased, curies	Price, $/curie
0–10,000	0.50
10,001–50,000	0.45
50,001–200,000	0.35
Over 200,000	0.125

The proposed price increase was canceled after the sales rate for the radioisotope increased rapidly and production and distribution costs decreased as a result of process improvements. The new processing techniques will enable existing AEC facilities to recover full costs at capacity production. Other factors include evidence of substantial private investments in research, development, and applications of ^{137}Cs and evaluation of public comments following publication of the proposed price increase. Future considerations of ^{137}Cs prices will take into account full costs; growth of market demand; extent of private research, development, and applications investment; and the possibilities for private separation and marketing of the material.

Cesium-137 is obtained from the waste solutions left after spent reactor fuels are chemically processed to recover unused uranium.

Carbon-13

Prices of ^{13}C of 90% or greater isotopic purity have been decreased from $4400 to $685 per gram by Mound Laboratory, Miamisburg, Ohio.

ACCELERATORS

Radiation Dynamics, Inc., of Westbury, L. I., N. Y. (11590) is offering its Dynacote industrial electron accelerators for inducing cross-linking, polymerization, and grafting in organic monomers. The penetration of the accelerator beam varies from 17 mils for the 300-kv machine to 32 mils for the 500-kv. Radiation doses from 1 to 10 Mrads can be delivered to products at a throughput rate of from 2.5 thousand to 100 thousand sq ft/hr or from 2 hundred to 9 thousand lb/hr.

COURSES AND CONFERENCES

The International Atomic Energy Agency (IAEA) and the Food and Agriculture Organization (FAO) sponsored an international training course on food-irradiation technology and techniques at the Massachusetts Institute of Technology in Cambridge, which ran from June 17 through July 25, 1969. About 20 scientists from a similar number of countries attended the 6-week session. The same two agencies sponsored an 8-week training session on the use of radioisotopes and radiation in entomology at the University of Florida, in Gainesville, from Oct. 6 through Nov. 28, 1969. The U. S. Atomic Energy Commission participated in both courses.

Scientists from some 20 countries presented papers at a symposium on plant mutations at Washington State University, in Pullman, on July 14–18, 1969. This symposium, on the nature, induction, and utilization of mutations in plants, reviewed important progress in this field since 1964, when a similar symposium was held at FAO headquarters in Rome. The proceedings will be published by the sponsoring agency, the IAEA, in about 6 months.

A 5-day panel on the analytical control of radiopharmaceuticals met in Vienna from July 7 to 11. Sponsored by the IAEA, this panel marked the Agency's entry into this branch of nuclear medicine. There were 27 participants from 15 countries and three international organizations, among them being producers of radiopharmaceuticals, users, and technical experts.

NBS OFFERS RADIOACTIVITY STANDARDS

The National Bureau of Standards (NBS) certified and issued four new radioactivity standards during the past year:

^{113}Sn–^{113m}In: The equilibrium product, as the chloride in 2.5*M* HCl, is supplied in a sealed ampoule containing 5.279 ± 0.009 g of solution. ($85)

^{226}Ra: Developed primarily for radon work, this standard consists of 20.598 ± 0.017 g of an HCl solution of ^{226}Ra with 0.2 wt.% $BaCl_2$ carrier. ($76)

^{228}Th–^{208}Tl: The point source is made from ^{228}Th, in equilibrium with its daughters, deposited as the nitrate on 0.019-cm-thick gold foil cemented to another similar gold foil, and all sandwiched between two double layers of 0.036-cm-thick polyurethane film. ($93)

^{238}Pu: The practically weightless ^{238}Pu source is electroplated onto a 0.010-cm-thick platinum foil, which is cemented to a Monel disk 2.5 cm in diameter by 0.16 cm thick. ($78)

These standards may be bought, for the prices indicated, from the Office of Standard Reference Materials, Room B308, Chemistry Building, National Bureau of Standards, Washington, D. C. 20234.

ORNL STABLE-ISOTOPE INVENTORY

Recent additions to the Oak Ridge National Laboratory (ORNL) enriched-stable-isotope saleable inventory include the following:

Isotope	Purity, %	Amount available, g
^{58}Fe	66.2	2.9
^{58}Fe	82.5	7.1
^{67}Zn	89.5	0.5
^{98}Mo	97.0	116
^{114}Cd	98.6	15.0

In addition to the regular inventory, ORNL maintains a rather large research-materials collection; for further information regarding availability from this inventory, write Isotopes Sales Department, Oak Ridge National Laboratory, P. O. Box X, Bldg. 3037, Oak Ridge, Tenn. 37830. (PSB)

MOUND LABORATORY PUBLISHES STABLE-ISOTOPES CATALOG

Mound Laboratory, Miamisburg, Ohio 45342, has published a second revision of their Stable Gaseous Isotopes and Carbon-13 Catalog. Entitled *Stable Isotopes*, it lists higher purities and revised prices for isotopes of He, Ne, Ar, Kr, Xe, and C and includes information on shipping containers and analytical services.

Mound Laboratory is operated by the Monsanto Research Corporation, a subsidiary of Monsanto Corporation, for the U. S. Atomic Energy Commission.

AEC Activities

AGREEMENTS WITH STATES

The AEC has approved proposals by North Dakota and South Carolina under which the states will assume part of AEC's regulatory authority over the use of radioactive materials in these states. The transfer of responsibility will include licensing, rule making, and enforcement in the uses of radioisotopes, the source materials uranium and thorium, and small amounts of fissionable materials. There are about 40 licensees in North Dakota and 115 in South Carolina for the use of radioactive materials. The state health departments are responsible for administering the radiation-control program within the states, which will cover the use of radioactive material as well as X-ray machines and other sources of radiation not regulated by AEC. The AEC has found these states' proposed radiation-control programs to be compatible with the AEC program and adequate to protect public health and safety.

These are the 20th and 21st agreement states, the others that have been approved being Alabama, Arizona, Arkansas, California, Colorado, Florida, Idaho, Kansas, Kentucky, Louisiana, Mississippi, Nebraska, New Hampshire, New York, North Carolina, Oregon, Tennessee, Texas, and Washington.

SANDERS NUCLEAR CORPORATION CONTRACT EXTENDED

The AEC has extended its $280 thousand contract with the Sanders Nuclear Corporation of Nashua, N. H., for another year. The contract is for investiga-

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tion of the use of ^{170}Tm oxide as a fuel for isotopic power devices. As part of the AEC's general program to investigate radioisotopes that show promise as power sources, the Sanders study is to define operating limits for fuel capsules of various designs, select appropriate test procedures, and measure and test the properties and performance of thulium oxides.

ATOMIC BATTERY BEGINS 9th YEAR OF SPACE OPERATION

The SNAP-3A (Systems for Nuclear Auxiliary Power), a drum-shaped grapefruit-sized atomic battery powered with ^{238}Pu, began its ninth year in orbit on June 29, 1969, although designed for only a 5-year life. This generator, developed by the AEC, is supplementing the power from solar cells on the Navy's oldest operating navigational satellite in space and the first to carry a nuclear power supply. On its eighth anniversary, the satellite had circled the earth 40,530 times and traveled more than 10^9 miles.

It is now operated on command from The Johns Hopkins Applied Physics Laboratory, Howard County, Md., and still beams information "loud and clear." This satellite was a forerunner of the Navy satellite navigation system, now used by the Fleet and by scientific and commercial ships.

April 1969 saw the first civilian use of atomic energy in space with the launching of NASA's Nimbus-3 weather satellite, which carries two nuclear generators as supplementary power to solar cells. This combined power is resulting in the acquisition of data of such significance that it has been characterized as "unprecedented" by the scientific community. When Apollo 12, the second manned lunar landing mission, reached the moon, an atomic-fueled generator was placed on the lunar surface, where it will provide the power for a package of instruments.

NUCLEAR GENERATORS IMPLANTED IN PACIFIC FOR OCEAN EXPLORATION

Two compact 10-watt power generators—SNAP-21, fueled with ^{90}Sr—have been implanted, for the AEC, for long-term testing by the Navy Undersea Research and Development Center, Pasadena, Calif., and the Naval Radiological Defense Laboratory, San Francisco, off the coast of San Clemente Island as part of a joint AEC-Navy program. A third unit is to be implanted later.

These generators were developed by the AEC to meet an increasing need for long-endurance and highly reliable power sources for underwater navigational aids, sonar beacons, seismological stations, and general-purpose oceanographic and ocean-engineering application in the ocean at depths as great as 23 thousand ft. They are designed to provide uninterrupted electric power for 5 years or more; conventional power sources are generally limited to lives of less than 6 months. The San Clemente test is expected to last 1 to 2 years, during which time the highly instrumented prototype units will be monitored continuously to determine their long-term behavior in an ocean environment. Initial tests will be at 60 and 300 ft to subject the nuclear devices to marine growth generally found at 500 ft or less. Deep-water tests are planned following successful completion of the shallow-water tests. The units will eventually be recovered for laboratory analysis and evaluation.

The power units were developed for the AEC by the 3M Company, Space and Defense Products Department, St. Paul, Minn. The Linde Division of Union Carbide Corp., Buffalo, N. Y., provided the high-temperature vacuum insulation subassembly as principal subcontractor to the 3M Company. The ^{90}Sr fuel was provided by the AEC's Hanford Plant, near Richland, Wash., and recovered by the Oak Ridge (Tenn.) National Laboratory from reactor-fuel-processing waste. The fuel in the unit is encapsulated in a heavy-walled nickel-alloy cylinder capable of containing the fuel under normal and accident operating conditions for hundreds of years. An extensive safety-test program has been conducted to demonstrate complete integrity of the fuel container under all handling, transportation, implantation, operating, and accident conditions. Each power package is 16 in. in diameter, 28 in. high, and weighs about 700 lb, including shielding and pressure vessel.

Deployment of these nuclear power units represents the culmination of the initial phase of the AEC program to develop a second generation of highly reliable, long-endurance, and economic radioisotope power sources for terrestrial and marine applications.

CHANGES IN THE *Code of Federal Regulations*

To keep our readers up to date on changes in rules and regulations that might affect their use of radioisotopes, *Isotopes and Radiation Technology* publishes the following table:

Table 1 Changes in the *Code of Federal Regulations*

Proposed effect	Number of part to be changed*	Date published	Status
To exempt certain small quantities of by-product material now generally licensed, to revise radioisotope listing, and to conform to Appendix C of Part 20	10 CFR 20 10 CFR 30 10 CFR 31 10 CFR 32 10 CFR 35	8-10-68	Comment period has expired; under staff consideration
To exempt from licensing the use of small quantities of ^{3}H, ^{60}Co, ^{13}N, ^{85}Kr, ^{137}Cs, and ^{147}Pm in electron tubes and to revoke the general license for spark-gap tubes containing some of these radioisotopes	10 CFR 30 10 CFR 31 10 CFR 32	11-14-68	Rule became effective May 2, 1969
To exempt self-luminous products containing ^{3}H, ^{85}Kr, or ^{147}Pm from further regulatory control if they meet specific safety standards; toys and adornments would be excluded from the class exemptions	10 CFR 30 10 CFR 32	6-21-68	Rule became effective June 20, 1969
To exempt from licensing the use of radioisotopes in smoke and other gas and aerosol detectors designed to protect life or property from fires and airborne hazards, provided such detectors meet specific safety standards	10 CFR 30 10 CFR 32	11-1-68	Rule became effective May 2, 1969
To issue a general license for use of ^{125}I or ^{131}I in amounts of 10 μc or less for in vitro clinical or laboratory tests	10 CFR 31 10 CFR 32	4-26-68	Rule became effective Jan. 13, 1969
To amend the labeling requirements for luminous aircraft-safety devices used under general license	10 CFR 32	6-5-68	Rule became effective Dec. 7, 1968
To bring AEC regulations on safe transportation of radioactive materials into conformity with the Oct. 4, 1968, revision of Department of Transportation regulations	10 CFR 71		Rule became effective Dec. 31, 1968
To redefine the category of products containing radioactive materials over whose transfer the AEC retains jurisdiction in an agreement state	10 CFR 150	2-24-68	Rule became effective May 2, 1969

*Title 10, *Code of Federal Regulations.*

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FOOD-IRRADIATION PROGRAM NOT DEFUNCT

The AEC and the Army hope to have a more extensive food-irradiation program than was anticipated after adverse Food and Drug Administration (FDA) action in 1968 and a budget cut in 1969. Upset by the AEC FY 1970 budget's lack of provision for the food-irradiation program, the Joint Committee on Atomic Energy requested an additional $750 thousand for the Division of Isotopes Development and a reassignment of $700 thousand to the Division of Biology and Medicine. Favorable Congressional appropriation action is expected.

The FDA rejected the Army's ham petition and withdrew its approval of irradiated bacon in 1968. This affected not only the Army's program but also the AEC's, which was depending on the Army's high-dose wholesomeness data to support its own petitions to the FDA for approval of low-dose irradiated foods. The FDA has now defined its requirements more precisely, and the Army has submitted a research program on irradiated canned ham. If FDA action is favorable, the Army will go ahead with 3-year animal-feeding studies on ham. Industry will be asked to bid on making the ham-wholesomeness studies that the Army cannot handle. Wholesomeness studies on chicken, beef, pork, hot dogs, luncheon meat, lamb, turkey, shellfish, finfish, and codfish and salmon cakes are expected to follow.

Plans for the Division of Biology and Medicine's $700 thousand reassigned funds are not yet complete but are expected to include animal-feeding studies on haddock, cod, and shrimp.

WPC FLOORING IN AEC BUILDING

The AEC has replaced the 11-year-old bluestone flooring in the lobby of its Germantown headquarters with Gammapar wood-plastic combination (WPC) parquet. The maintenance cost for this material is sufficiently lower than that of other materials considered to offset the somewhat higher initial cost. The supplier of the material is American Novawood.

FILM ON WOOD–PLASTIC MATERIALS

"Atomic Revolution in Wood" is a new, popular-level motion picture outlining man-made improvements to wood products with the use of nuclear energy. The 23-min color film, produced for the AEC by the Army Pictorial Center, discusses the fusing of wood and plastic by irradiation into a substance that has greatly improved hardness, abrasion resistance, and durability as well as all the aesthetic appeal of wood. The film traces the development of radiation-processed wood-plastic materials from initial AEC research and development through commercial production by several companies.

The movie may be borrowed free of charge, except for return postage, from the Motion Picture Film Library, U. S. Atomic Energy Commission, P. O. Box E, Oak Ridge Operations, Oak Ridge, Tenn. 37830.

General

PHO/GAMMA CAMERA POPULAR

Nuclear-Chicago Corp. is making extensive sales of the Pho/Gamma Camera, which is now in use in 5% of U. S. hospitals. This camera is the commercial version of the Anger camera, over which Nuclear-Chicago and Picker Corp. have had patent difficulties. The camera is used in nuclear medicine for showing the location of tracer amounts of radioisotopes used in diagnosis.

GAMMA RADIOGRAPHY STUDIED

Nondestructive testing of aircraft with gamma radiation to determine the amount of bullet protection provided by an aircraft's structure is a project of the Cornell Aeronautical Laboratory in Buffalo, N. Y., for the Air Force. A correlation between the gamma-radiation attenuation by a plane's structure and its ballistic stopping power will allow determining the best armor protection with least weight.

RADIATION TECHNOLOGY'S ACTIVITIES

Radiation Technology, Inc., is irradiating tons of enzymes at its St. Hilaire, Quebec, facility to decrease the spore count to preset levels, with only minor decrease in enzyme activity. The method is considered

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competitive with filtration, but the throughput is higher. Costs vary with the enzyme bulk density, the throughput, and the package geometry and can be less than 10¢ per pound.

The St. Hilaire facility is also of interest for catalyzing the polymerization of latex emulsion paints and adhesives. A continuous production facility to produce latex formulations is a prime objective because both polyvinyl acetate and polystyrene can be produced with narrower particle-size ranges and a more uniform molecular-weight distribution than can conventionally produced polymers.

Radiation Technology developed a process at this facility for inducing crease resistance in woolens. The results of about 150 experiments on wool demonstrated that a crease recovery angle comparable favorably to that in control materials could be made. Research on synthesis of organometallic compounds is planned.

PICKER TO SUPPLY RADIOPHARMACEUTICALS

A new company has been formed by Picker Corp. of White Plains, N. Y., and American Hoechst Corp. of Bridgewater, N. J. (subsidiary of Farbwerke Hoechst AG of Frankfurt, Germany) to supply radiopharmaceuticals in North America. The new company is located in Bedford, Mass., and is headed by Dr. H. Huckel, a vice-president of American Hoechst.

RADIATION MACHINERY CORP. REPORTS DEVELOPMENTS

Radiation Machinery Corp. expects more than $5 million in revenue for the fiscal year ending June 30, 1970. Its $430 thousand earnings of the previous year came from sales of educational and industrial irradiators. Current earnings will come from these plus sale of wood-plastic flooring.

The company's Hanover, N. J., wood-plastic flooring plant has a 5-Mc ^{60}Co radiation source, and its present annual capacity of 3 million sq ft of flooring is expandable to 18 million sq ft. It is planning a new plant in West Virginia, which will have a 10-Mc ^{60}Co source and an annual capacity of 40 million sq ft. George R. Dietz, formerly of AEC's Division of Isotopes Development and now Assistant Vice-President for Operations at Radiation Machinery, is in charge of production of wood-plastic flooring and of irradiation facilities. The company is also planning to build a plant in Puerto Rico for production of irradiated concrete plastics.

Radiation Machinery has bought 0.1 Mc of ^{60}Co from Neutron Products, Inc., for research use but has ordered 1 Mc from Atomic Energy of Canada Limited in preference to U. S. sources, which it felt either could not meet delivery schedules or had a price that was "way too high."

BETA GAGE RESEARCH AND DEVELOPMENT

The U. S. Army Cincinnati Procurement Agency, 550 Main St., Cincinnati, Ohio 45202, is starting a research and development program for design and assembly of a point-source beta gage system for use in quantitative, nondestructive measurement of weight loss of fabrics due to wear. The system must be able to detect wear or weight loss from the time it starts through the full weight range. No instrument design is considered necessary.

AECL SUPPLIES ^{60}Co SOURCES

AECL Commercial Products has fabricated a ^{60}Co facility for the new university hospital in London, Ontario. It previously fabricated two for European customers and one for a North American firm for sterilization of medical supplies. Each of the three units can accommodate sources of a million curies, but initial loadings are only a fraction of the total capacity.

Rental programs have become an important part of AECL marketing practices. Cobalt-60 research irradiators are being made available on a lease basis, which will make it easier for research and development laboratories in the United States and Canada to revise radiation-research projects (or introduce new ones) that have been curtailed for lack of capital funds. Four standard models are being offered, although a new "mouse irradiator" may be offered in the future. Terms include an option to purchase, full warranty and guarantee, maintenance, and servicing.

^{60}Co NAVIGATION LIGHT TO BE TESTED

A prototype 5-watt ^{60}Co power source is being evaluated by Canada's Department of Transport as a navigation light on the St. Lawrence Seaway system. AECL feels that ^{60}Co has the edge over ^{90}Sr in terms of guaranteed availability and a predictable price struc-

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ture. Cobalt-60 has a reasonable half-life, an excellent fuel form for mechanical stability and heat transfer, and a lower toxicity hazard than ^{90}Sr. Partially decayed ^{60}Co can be used for other purposes, such as industrial gamma irradiation, without reprocessing, and its shielding requirements are little more than those for ^{90}Sr for power outputs exceeding a few hundred thermal watts.

A proof-of-principle ^{60}Co-fueled light was the MAPLE 1A (Minor Atomic Prolonged Life Equipment). The MAPLE 1B, which is being used in the test, is an improved version of the 1A. A first-generation MAPLE should cost less than $20 thousand but is still considered expensive as a purchase to be amortized over a 5-year working life. AECL believes that, by allowing a reasonable depreciation of the hardware capital cost (if it can be reused) and calculating fuel costs on the basis of actual decay, much lower unit power costs can be achieved.

1968 RADIOACTIVE RADIATION SALES

Total atomic energy products shipments in 1968—including those to U. S. government agencies and exports of selected important products made in privately owned industries—were valued at $510.5 million. Of this total, $16.9 million represented radiation sources and other radioactive materials produced from purchased isotopes and $8.1 million represented commercial irradiation services and processing of sources and special nuclear materials.

DONALD W. DOUGLAS LABORATORIES DEVELOP LOW-LEVEL NUCLEAR BATTERIES

Donald W. Douglas Laboratories of McDonnell-Douglas Astronautics Company's Western Division have recently reported the development of prototype nuclear batteries in the 10-μw to 1-watt range. These include two types of devices: a thermal battery with a thermionic converter powered by nuclear heating [TM, Isomite (Fig. 1)] and an electron-voltaic battery that derives its power from the interactions of nuclear decay particles [TM, Betacel (Fig. 2)]. The chief targets for these devices are telemetry equipment (1 watt or less), hearing aids (1 mw), heart pacemakers (100 μw), and electric watches (10 μw).

The Betacels are convenient for multicell packaging, giving an almost limitless number of possible outputs through variation in size of the wafer and the series-parallel connections of the cells. The Isomite batteries, on the other hand, operate in a power area previously considered impractical for thermionic energy conversion.

Although nuclear batteries are not suited to all battery applications (the desirability in a given application must be evaluated for that application in terms

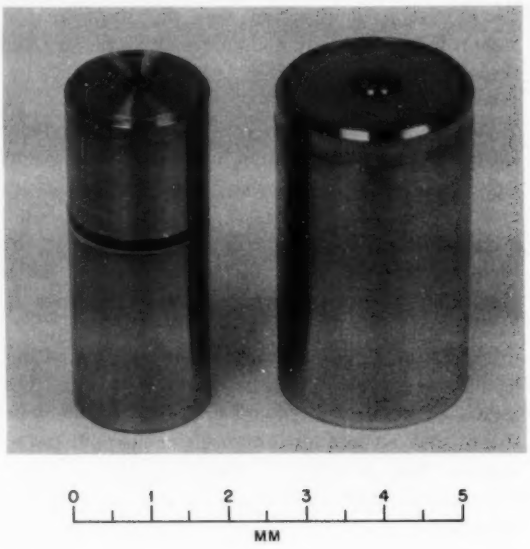

Fig. 1 Isomite battery—a miniature radioisotope-fueled thermionic converter.

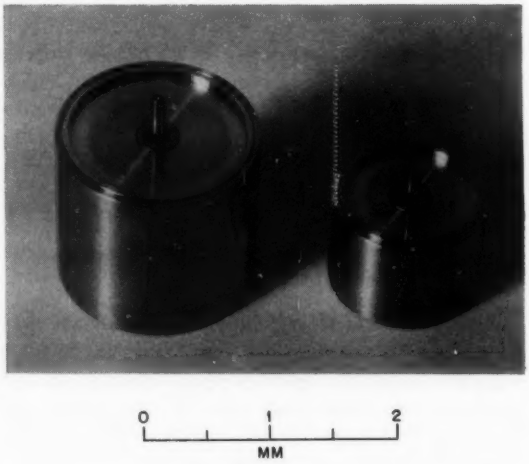

Fig. 2 Betacel battery—a miniature radioisotope-fueled betavoltaic power source.

of size, weight, lifetime, and cost), certain advantages are noted:

1. Their energy-storage potential is superior to conventional batteries, and their delivered energy per weight is 10 to 100 times that of chemical batteries.
2. Their lifetimes can be 1000 times or more those of other batteries.
3. Their reliability is potentially far superior to that of other batteries.
4. For long lifetimes, where recharging is necessary for conventional batteries, the unit cost of delivered energy can be lower than for other batteries.

However, it must be recognized that there are also some drawbacks associated with nuclear batteries:

1. The initial cost is generally higher than for conventional batteries.
2. For most practical applications, the potential very high energy/weight ratio is not possible because of shielding requirements.
3. The radioisotopes used in the nuclear batteries are inherently more hazardous than the materials in nonnuclear batteries.

TRENHOLME BECOMES CONSULTANT

William N. Trenholme has resigned his position as Director, state of Arizona, Atomic Energy Commission, to become a consultant. His consulting services are offered to business, medical, educational, and governmental organizations in the areas of advice on procurement of equipment for use of radioisotopes and other radiation sources, assistance in training personnel and meeting government regulations, and advice on nonclassified nuclear and electronic developments in the United States and their possible application to the needs of foreign countries. His address is 40 East Thomas Road, Suite 118, Phoenix, Ariz. 85012 (Phone: 602-265-2198).

AECL COMMERCIAL PRODUCTS RECORDS BEST SALES YEAR YET*

Isotope and radiation-equipment sales by AECL's Commercial Products group finally exceeded $10 million, an increase of 11% over the previous year. The actual figure was $10.3 million for the fiscal year ending Mar. 31, 1969. No turn-key industrial irradiators were delivered in that year, which augurs well for income in the 1969-1970 period since several are in production and a large number of new orders are in hand. The 10-million mark was nudged in the cumulative total of curies shipped since AECL began irradiating cobalt and other materials at Chalk River. Last year's contribution was 1.8 million curies.

AECL is still one of the big three in North America's ^{60}Co market. This is now only about 2 million curies a year, but one guess is that it will be close to 10 million curies by 1975. The only problem for suppliers like AECL, General Electric, and Neutron Products is that power reactors will be capable of producing several times that amount.

AECL's annual report gave considerable attention to gamma irradiation of grain and vegetable seeds. For the 1969 crop year, a sizable expansion of the program carried out in cooperation with the federal Department of Agriculture is planned. Field corn and barley will be included with the sweet corn, lettuce, tomatoes, and eggplant for testing in Ottawa, and additional tests will be initiated at Kemptville, Ontario; Winnipeg and Beaverlodge, Saskatchewan; and Lethbridge, Alberta. The tests in western Canada will include wheat, oats, barley, flax, forage grasses, and some vegetables. The short growing season of the northern region at Beaverlodge makes possible an evaluation of the possibility of the technique for increasing yields in severe climates. A few hundred rads is a normal dosage, and there is evidence that, in some varieties at least, significant growth stimulation occurs.

At least one Canadian food processor has been following up some of the work done by the U. S. Army Laboratories in Natick, Mass. Salada Foods Limited (Toronto) has been granted a patent covering an irradiation process for dehydrated potatoes which involves immersing the potatoes in a bath of cryogenic nitrogen and irradiation at dosages of up to 10 Mrads. By bringing temperatures down, the researchers were able to get products superior in texture, flavor, and nutrition. Salada did some of their work at Natick. Like the U. S. Army, Salada has an engineering problem to consider. The potatoes must be kept at cryogenic temperatures while they pass through the ^{60}Co irradiator maze. The economics of using irradiation to reduce rehydration times of dehydrated products have yet to be assessed.

HIGH VOLTAGE INTEREST IN JAPANESE POLYMER

As the result of an agreement between the Sekisui Chemical Company Limited of Osaka, Japan, and High

*From *Nuclear Canada*, 8(7): 12-13 (July-Aug. 1969).

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Voltage Engineering Corp., of Burlington, Mass., a new company has been formed to produce polyethylene foam products that are cross-linked by irradiation. The new company, Voltek, Inc., expects to start operations early in 1970. In the meantime, the Japanese products are being imported in limited amounts to introduce consumers to the materials, which are sold in Japan as "Softlon" and "Eftlon." These are closed-cell polyethylene materials and feature good engineering properties as well as a very smooth skin. They are easily cut, sliced, and punched, and they can be combined with each other, with other foams, and with paper and textiles. They do not absorb water, they have good insulation properties, and they are resistant to chemicals.

ACS NUCLEAR CHEMISTRY AND TECHNOLOGY DIVISION ANNOUNCES EDUCATIONAL AWARDS

The Division of Nuclear Chemistry and Technology of the American Chemical Society is offering up to two annual awards of $500 each to those undergraduates, among nominees of their respective faculties, whose completed nuclear or nuclear-oriented projects in chemistry or chemical technology are judged by the Division to have best embodied ingenuity, novelty, and potential usefulness. Nominations, and project descriptions written by the nominees, must be in the hands of the Division Secretary by May 31 of each year. Winners will be notified through the nominating faculty members by July 31, and the awards will be appropriately announced at the National Meeting the following fall. A simple definition of "nuclear-oriented" will be used: centrally involving nuclear phenomena, their effects, and their measurement. Examples: nuclear chemistry, radiation chemistry, chemistry of isotopically labeled compounds, tracer applications, chemistry of interest to nuclear energy programs, and effects of isotopic differences even when no radioactivity is involved.

The Division is offering this award to publicize the nuclear aspects of chemistry and chemical engineering. An equally important objective is a more widespread realization among undergraduates of the benefits to society and the help for solving society's problems which reside in understanding and exploitation of nuclear phenomena.

Additional information may be obtained from:

Dr. Donald C. Stewart
Argonne National Laboratory
9700 S. Cass Avenue
Argonne, Ill. 60439

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